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MARTIN MARIETTA

INVESTIGATION OF A URANIUM HEXAFLUORIDE RELEASE INCIDENT ON SEPTEMBER 17, 1975, IN THE K-1423 TOLL ENRICHMENT FACILITY

Oak Ridge Gaseous Diffusion Plant

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Title/Subject INVESTIGATION OF A URANIUM HEXA-FLUORIDE RELEASE INCIDENT ON SEPTEMBER 17, 1975, IN THE K-1425 TOLL ENRICHMENT FACILITY.

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Title: "Inventory of a Uranium Hexafluoride Release Incident of September 17, 1975, in the K-1423 Toll Enrichment Facility"
Authors: A. J. Legeay, et al.
Abstract: On September 17, 1975 at the ORGDP Toll Enrichment Facility a 30A cylinder, having just been filled with 4,887 lbs of liquid UF6 enriched to 3.28% uranium-235, an explosion ocurred inside the cylinder. Smoke emerged from a crack in the cylinder wall and from a crack in the valve body. The release was controlled within ten minutes and stopped within 20 munutes subsequent cylinder weighings indicated the release of approximately 18 lb fo UF6; 2.3 lb of the material relased was recovered.
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Report K/P-6197, Rev. 1, is the same as K-P-6197 except that the names of employees have been deleted.

INVESTIGATION OF A URANIUM HEXAFLUORIDE RELEASE INCIDENT ON SEPTEMBER 17, 1975 IN THE K-1423 TOLL ENRICHMENT FACILITY (U)

Investigating Committee:

A. J. Legeay, Chairman
L. W. Anderson
J. W. Arendt
J. C. Bailey
E. J. Barber
J. C. Barton
T. B. Bomar
O. L. Calvert
H. R. Dyer
J. Dykstra
E. Y. Kimmerly
M. E. Mitchell
R. L. Newton
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UNION CARBIDE CORPORATION
NUCLEAR DIVISION

Oak Ridge Gaseous Diffusion Plant
Oak Ridge, Tennessee

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INTRODUCTION

An explosion occurred in a 30A 2-1/2-ton UF_6 cylinder at the K-1423 Toll Enrichment Facility (Figure 1) on September 17, 1975, cracking the cylinder and resulting in the release of about 18 lb of UF_6. The damaged cylinder was one of 10 cylinders received by ORGDP from an enriching service customer on March 19, 1975. These cylinders were cleaned and inspected at ORGDP, returned to the cylinder manufacturer for inspection and hydrostatic testing, then reinspected at ORGDP, valves installed and leak tested, and evacuated prior to filling with enriched UF_6 at the Toll Enrichment Facility.

The following committee was appointed to investigate the incident:

A. J. Legeay, Chairman - Operations
L. W. Anderson, QA Coordination
J. W. Arendt, Laboratory
J. C. Bailey, Health Physics
E. J. Barber, Development
J. C. Barton, Laboratory
T. B. Bomar, Safety
O. L. Calvert, Operations
H. R. Dyer, Criticality Safety
J. Dykstra, Operations
E. Y. Kimmerly, Equipment Test and Inspection
M. E. Mitchell, Environmental Management
R. L. Newton, Shift Operations
J. W. Pickel, Maintenance
K. T. Ziehlke, Metallurgical Evaluation

The committee purpose was to determine the cause of the incident, thoroughly evaluate procedures and practices and recommend necessary changes to preclude recurrence of a similar incident in the future. This report presents the investigations carried out by the committee and their conclusions and recommendations based on findings.

PHOTO NO 72 920

Figure 1

K-1423 TOLL ENRICHMENT FACILITY

SUMMARY

On September 17, 1975 at approximately 1:30 p.m. at the ORGDP Toll Enrichment Facility a 30A cylinder, having just been filled with 4887 lb of liquid UF_6 enriched to 3.28% uranium-235, was transported by forklift to the concrete storage pad outside K-1423. Two operators, one at each end of the cylinder, were present to assist in disengaging the lifting hooks. After the cylinder was lowered to the pad, an explosion occurred in the cylinder as evidenced by a loud report and the outward bulging of both concave ends of the cylinder. Smoke began to emerge from a crack in the valve body and a crack in the cylinder wall at the opposite end. One operator incurred a minor injury when he was struck in the hand by the lifting hook as it was propelled away from the end of the cylinder. The other operator incurred a serious injury when he was struck on the knee by the identification plate which was ejected from the end of the cylinder. This injury was subsequently adjudged disabling.

The Emergency Squads responded promptly. The release was controlled within 10 minutes after the incident and stopped within 20 minutes. Subsequent cylinder weighings indicated the release of approximately 18 lb of UF_6; 2.3 lb of the material released was recovered.

The release did not result in any significant effect on the environment. Only the cylinder and the pad area immediately beneath the cylinder location required decontamination. There were no serious exposures of personnel to uranium materials.

Although initial gamma radiation readings indicated that no criticality was involved, precautions were taken in the event some unknown conditions such as cylinder integrity, cause of reaction, and weather might result in a criticality.

The estimated total cost attributable to the release amounts to $5,363 which consists of:

Material Loss	$2,235	(5.519 kg U as UF_6 x $404.93)
Decontamination	312	(Operator labor @$13.00/man-hour)
Replacement of Cylinder	800	
Cylinder Disposition	2,016	(Maintenance labor @$12.70/man-hour)
Total	$5,363	

Investigation has shown that the cause of the explosion in the cylinder was a reaction between UF_6 and a hydrocarbon and the source of the hydrocarbon was oil probably introduced into the cylinder as a result of a motor failure on the mechanical vacuum pump used to evacuate the cylinder prior to filling.

To prevent similar incidents in the future it is recommended that the evacuation as well as pressuring of cylinders in UF_6 service be performed with oil free equipment, e.g., evacuation by air ejectors and pressuring with oil free inert gas.

If other equipment such as oil lubricated pumps or compressors are used for evacuation or pressuring of cylinders, a safety analysis should be made to assure design of the equipment and its operation precludes the introduction of reactive contaminants to the cylinder.

The incident must be viewed as a "near miss" of a more serious incident in that the potential existed for complete separation of the cylinder head and the more serious consequences therefrom.

Investigation confirmed in most areas that the QA plans and related operating procedures and Job Safety Analyses existed, were adequate, and were followed. Areas were identified where plans are needed. Division management has established a schedule to be pursued with systematic follow-up to ensure that necessary improvements are made.

DISCUSSION

History of Cylinder N-8 Prior to September 17, 1975 Incident

Early in 1975, Transnuclear, Inc. requested decontamination services for ten NUKEM 30A UF_6 cylinders. These cylinders had been in UF_6 service since their fabrication in 1969 and were scheduled for the 5-yr periodic internal inspection and hydrostatic strength test by the fabricator, Columbiana Boiler Corporation, as specified in ANSI N14.1*. The ten cylinders, numbered N-1 through N-10, were received at K-1420 on March 20, 1975. The ten cylinders were cleaned at the K-1420 Decontamination Facility according to Chemical Operations Standard Operating Procedure 1410-5 during the period April 1 through April 8, 1975. The cylinder decontamination log indicates that Cylinder N-8 was cleaned with a water wash followed by three Trioxide washes, steaming, and drying on the April 4, 4-12 shift and the April 5, 8-4 shift, with final inspection on April 5. Rubber stoppers were installed in the 1-in. valve opening since the valves were not replaced after cleaning.

The ten NUKEM cylinders were shipped from K-1423 to Columbiana Boiler Corporation on April 30, 1975 as requested by Transnuclear, Inc. for installation of valve couplings, inspection and testing. The ten cylinders were returned to K-1423 from Columbiana on September 8, 1975. All of the cylinders were then transferred to the K-1420 Decontamination Facility on September 9, 1975 for internal inspection. Cylinder N-8 and eight other cylinders were inspected and approved for use. Cylinder N-6 was steam cleaned to remove a visible spot. The ten cylinders were transported on September 9-12, 1975 to the K-1401 Valve Shop for valve installation and air leak test as specified in ANSI N14.1.*

The routine sequence of operations performed in the K-1401 Valve Shop on UF_6 cylinders is detailed in Maintenance Procedure MEP-300. A brief summary of these operations includes the following:

1. Installation of a new valve;
2. Pressuring the newly valved cylinder to 100 psig with plant air for leak testing;
3. Air leak soap testing of valve by inspector;
4. Venting cylinder to atmospheric pressure; and
5. Evacuating cylinder to 27-in. Hg vacuum with a Kinney mechanical vacuum pump.

The exact details and timing of the sequence of Valve Shop operations could not be determined for the NUKEM cylinders. Review of logs and interviews with ten employees assigned to performing the cylinder work or repair of valve shop equipment were conducted by the Investigating Committee members to develop the probable chronology of events in the Valve Shop.

*ANSI N14.1 1971 American National Standard Packaging of Uranium Hexafluoride for Transport, pp 34 and 35.

Valves were installed in Cylinders N-2, N-5, N-7, and N-8 on September 9-10, 1975. These cylinders were also evacuated on the September 10, 4-12 shift. The motor of the Kinney mechanical vacuum pump used to evacuate the cylinders failed on this shift during the evacuation of either Cylinder N-7 or N-8. (See Illustration 1) When the pump motor started smoking, the evacuated cylinder was isolated from the pump by closing the evacuation manifold valve. The remaining 6 NUKEM cylinders were valved, pressured to 100 psig, leak tested, tagged as having acceptable valve leak rates, and vented to atmospheric pressure subsequent to the 4-12 shift on September 9-10.

There is no direct evidence that oil was introduced to any of the cylinders during the cylinder evacuation operation; however, significant quantities of oil have been demonstrated to be siphoned from the Kinney pump to an evacuated cylinder provided the cylinder valve and the evacuation manifold valve on the pump are open at the time the pump is shut down or fails. A summary of a test of the Kinney mechanical vacuum pump is presented in Appendix A.

The Valve Shop plant air header was checked to determine whether a significant quantity of oil was present. These tests conducted following the September 17 incident were negative. K-1401 Valve Shop personnel did not observe that oil was introduced to any of the NUKEM cylinders.

The cylinders were transported to the K-1423 Toll Enrichment Facility on September 11, 12, and 15. Final evacuation of those cylinders, which could not be completed at the valve shop because of the pump motor failure, was accomplished with the air ejector at K-1423. Tare weighing of the cylinders was completed at K-1423 on September 16. It was determined that Cylinder N-8 weighed 1390 lb. It should be noted that the previous tare weight on this cylinder was 1399 lb upon receipt on March 20, 1975. Since the cylinder had been decontaminated, painted, and valved between weighings, the weight loss was considered acceptable and did not indicate the presence of contaminants. The cylinder pressure was determined and recorded on all of the NUKEM cylinders as specified in the Toll Enrichment Facility procedure. A vacuum of 20-in. Hg was reported on Cylinder N-8.

The K-1423 Toll Enrichment personnel were interviewed to determine the sequence of cylinder handling operations involving the release incident. Information was also obtained from sources such as foremen's logs, print weight tickets, SS transfers, and inspection forms. It was determined from this information that 14-ton product Cylinder No. 9634 was filled with 26,452 lb of enriched UF_6 at 3.28% uranium-235 on August 22, 1974, at the K-413 Product Withdrawal Facility: This cylinder was heated in a K-1423 steam hood at 200° for homogenization and sampling on September 16, 1975. The content of this cylinder was scheduled for sampling and transfer to 5 NUKEM-owned 30A cylinders for the October 1975 shipment to Reactor Brennelemente Union in West Germany. The product was part of KES-250 totaling 20,760 kg uranium for Swedish State Power Board Ringhals-2 reactor. Early on the September 17, 8-4 shift, two liquid samples of UF_6 were withdrawn from Cylinder No. 9634 into 2S Cylinders Nos. 019 and 0329.

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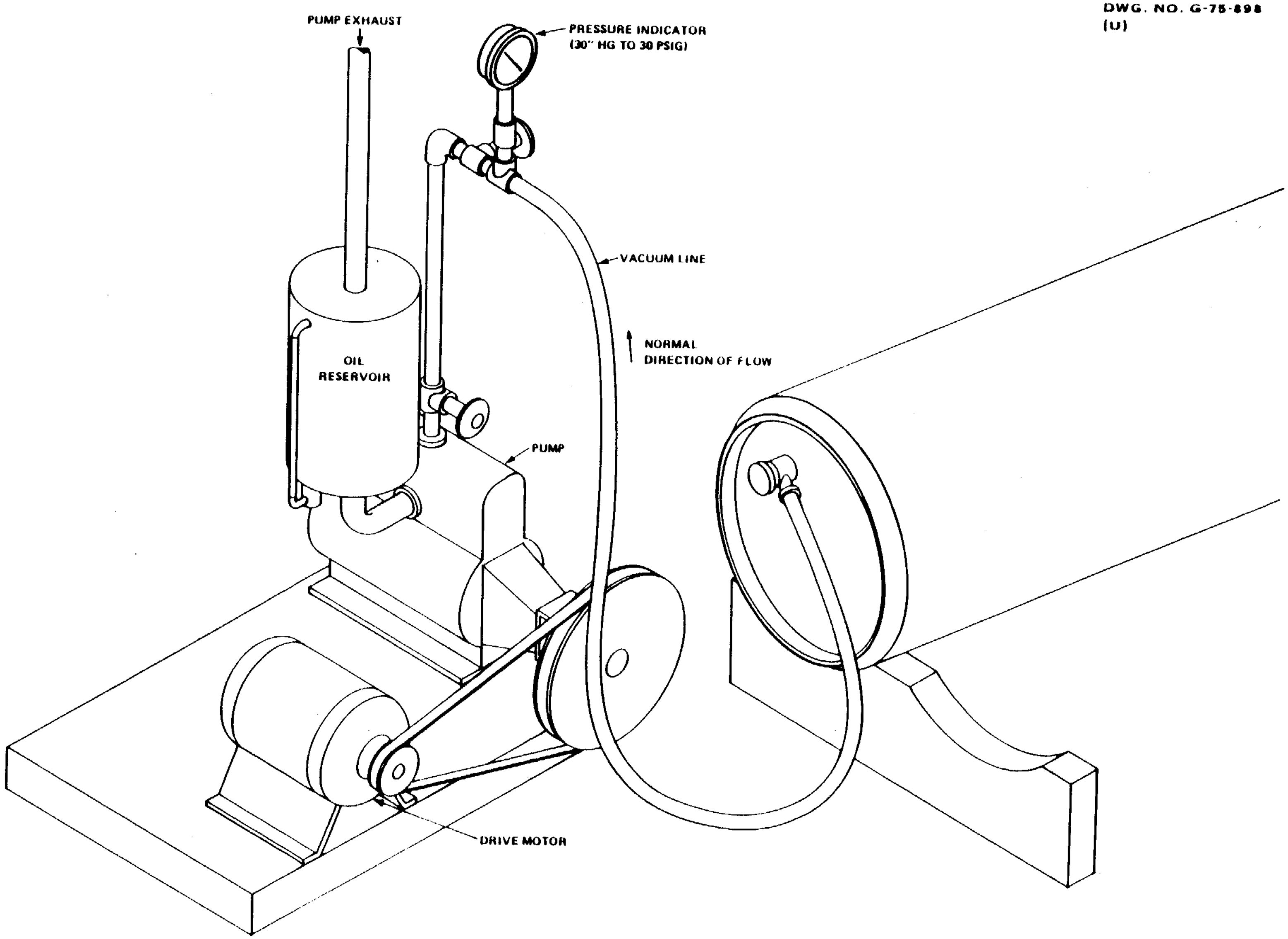

KINNEY VACUUM PUMP MODEL 8-8-10
'ILLUSTRATION 1

The product transfer operation data from the 14-ton Cylinder No. 9634 are presented below:

30A Cylinder No.	Weights, lb Gross	Tare	Net	Pressure In. Vac.	Transfer Time Start	Complete
N-1	5958	1320	4638	25	9:20 a.m.	10:35 a.m.
N-3	6214	1322	4892	25	10:40 a.m.	11:40 a.m.
N-8	6277	1390	4887	20	11:45 a.m.	1:30 p.m.

It should be noted that liquid transfer flow rate decreased as Cylinder N-8 approached the target fill weight. The cylinder was valved off from the transfer manifold and burped by valving into the evacuation manifold. In this operation, an air ejector is utilized to evacuate the manifold. Volatiles evacuated from the cylinder are passed through a cold trap and two alumina traps in series for collection of UF_6 prior to venting. After the single burping operation, filling of the cylinder was completed at a normal filling rate. Although the burping operation is not an everyday occurrence, occasionally such an evacuation is required to maintain the transfer rate as the final cylinders of a liquid transfer approach the fill limit. In the case of Cylinder N-8, the burping may have been required to evacuate volatile reaction products; however, later attempts to identify these reaction products in the cold trap and alumina were unsuccessful.

Description of Incident

Immediately after filling, Cylinder N-8 was moved by forklift to a storage area west of K-1423 for cooling (Figure 2). After the cylinder was lowered to the concrete storage pad, operators No.1 and No.2 proceeded to disengage the lifting fixture hooks from the cylinder. A single loud noise occurred, and the concave ends of the cylinder bulged and gas emerged from a crack in the valve body and a circumferential split of approximately 2 in. in length in the cylinder wall 2-1/2 in. from the opposite end of the cylinder (Figures 3 and 4). Operator No.1 was struck in the right hand by the lifting hook as it was propelled away from the cylinder; however, he was not seriously injured. Operator No.2 incurred a laceration of his left knee and was knocked to the ground when the metal identification plate was forcibly ejected from the opposite end of the cylinder.

Witnesses reported that the gas release from the damaged cylinder was projected in a southwesterly direction. The height of this plume was apparently about 25 ft above the ground; the bottom of the plume was about 3-4 ft above the ground and thus did not restrict visibility. Favorable metrological conditions (light westerly wind and light rainfall) prevented the rapid widespread dispersion of the plume and thus minimized emergency problems.

The Emergency squads reacted promptly (See Appendix B). The application of wet towels on the damaged cylinder valve and the crack in the cylinder wall controlled the release within 10 minutes after the incident.

PHOTO NO. 75-3000

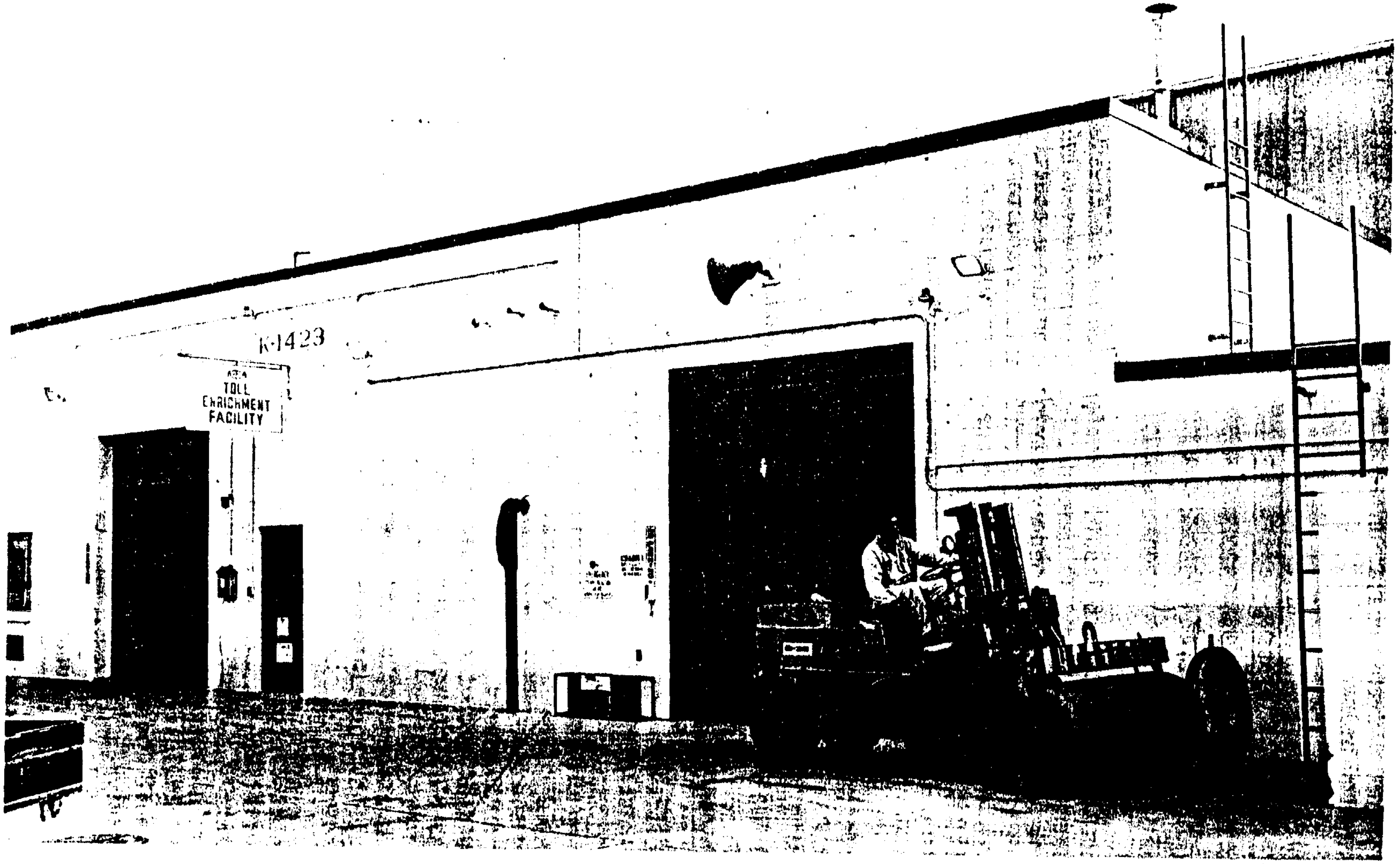

Figure 2

STORAGE AREA WEST SIDE K-1423

PHOTO NO. 75-2700

Figure 3
VALVE END OF DAMAGED CYLINDER

PHOTO NO. 75-2702

Figure 4

PLUG END OF DAMAGED CYLINDER

Refrigeration of the cylinder was provided by spraying CO_2 from a portable fire extinguisher, packing with dry ice, and directing a controlled stream of water from a 3/4 in. rubber hose onto the middle of the cylinder to avoid introduction of water to the cylinder ruptures. The damaged cylinder was allowed to cool for 4 days to condense and solidify the UF_6 contents prior to any recovery attempts. During this period, the area was isolated by barricades, and the cylinder was covered with tarps to protect it from rainfall.

Disposition of the Damaged Cylinder

Initial analytical investigation and metallurgical examination (Appendixes F and E) indicated the need to provide securement for the cylinder ends and secondary containment of cylinder contents prior to handling or transport. Steel end plates and tie rods were fabricated and installed (Figure 5). A lifting fixture was provided to handle the assembly (Figure 6) and secondary containment was provided with a modified converter shell (Figure 7). The secured cylinder was then transported to the cascade for vapor feeding of contents (Figure 8).

The condensed UF_6 was evacuated from the cylinder by sublimation at sub-atmospheric pressures. The cylinder was emptied without incident. The residue in the evacuated cylinder was sampled and the interior of the cylinder was inspected for visual damage.

Environmental Impact

In an effort to determine the effects of uranium and HF releases to off-site areas, the Environmental Management Group initiated sampling procedures immediately after being notified of the release. All sampling was under way by about 2:30 p.m. and continued for 24 hours. The results of these samples indicated that no uranium and very little, if any, fluorides escaped the ORGDP site. Since the time lapse between the release and the initiation of the atmospheric samples could have allowed for the gaseous plume to escape the plant boundary without being sampled, worst-case atmospheric dispersion calculations were made to determine maximum possible fence-line HF and uranium concentrations. The results of these calculations revealed that the HF concentration could not have exceeded 1.8 mg/m^3, which is about 90% of the recommended 8-hour exposure threshold limit and 7.2% of the threshold for physical discomfort. The uranium concentration at the fence could not, according to the dispersion calculation, have exceeded 5.5 mg/m^3. This concentration would have, when averaged over a period of 1 week, amounted to about 2.4% of the ERDA chemical toxicity limit and 2.0% of the alpha activity limit. Although the effects of such low concentration of uranium and HF on terrestrial vegetation for only 10 minutes, the duration of the release, are unknown, it appears that the released UF_6 had an insignificant impact on the surrounding environment. The environmental impact report is presented in Appendix C.

PHOTO NO. 75-3118

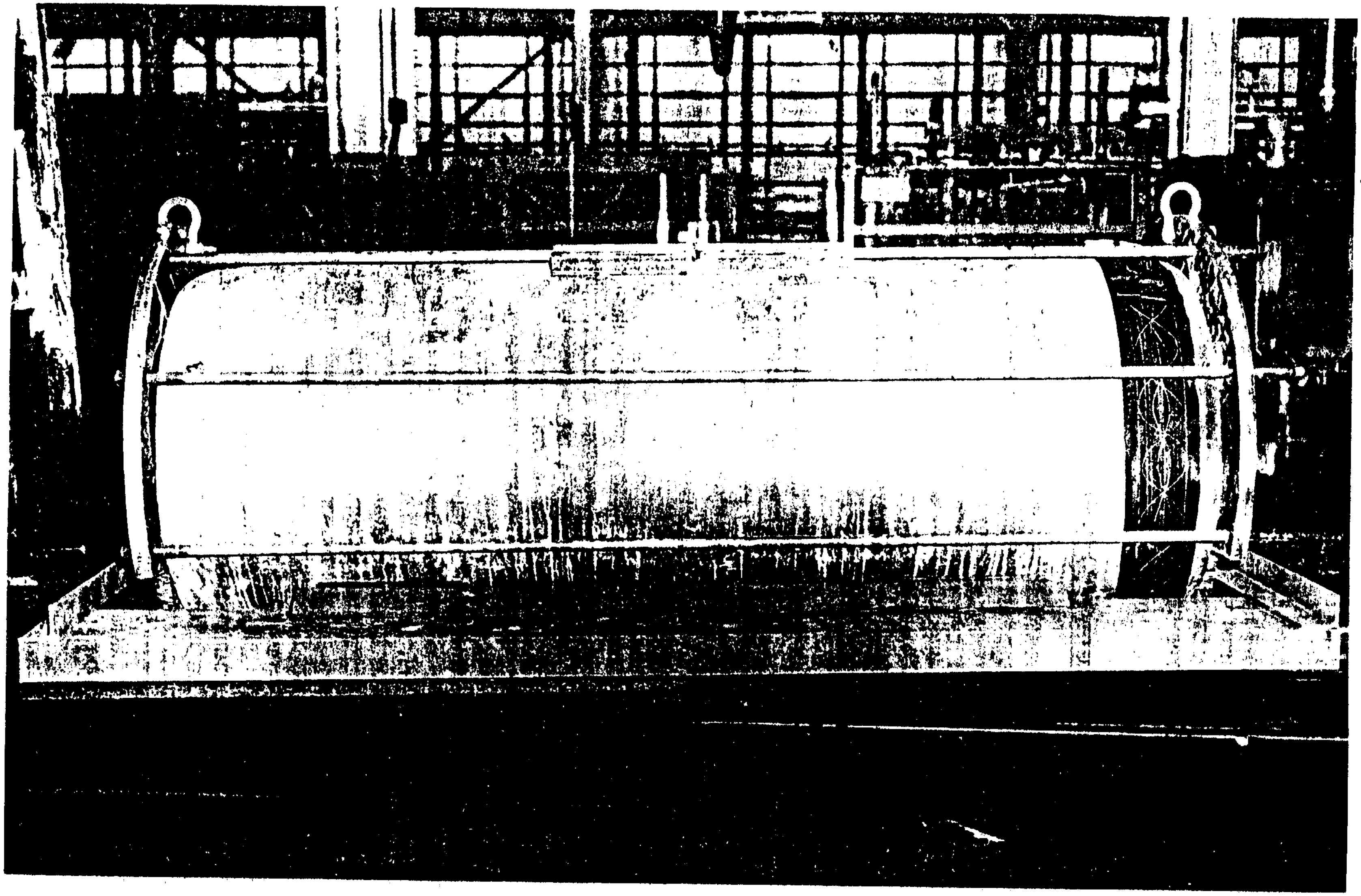

Figure 5

DAMAGED CYLINDER WITH END PLATES AND RODS

PHOTO NO. 75-3115

Figure 6
LIFTING FIXTURE FOR DAMAGED CYLINDER

PHOTO NO. 75 3116

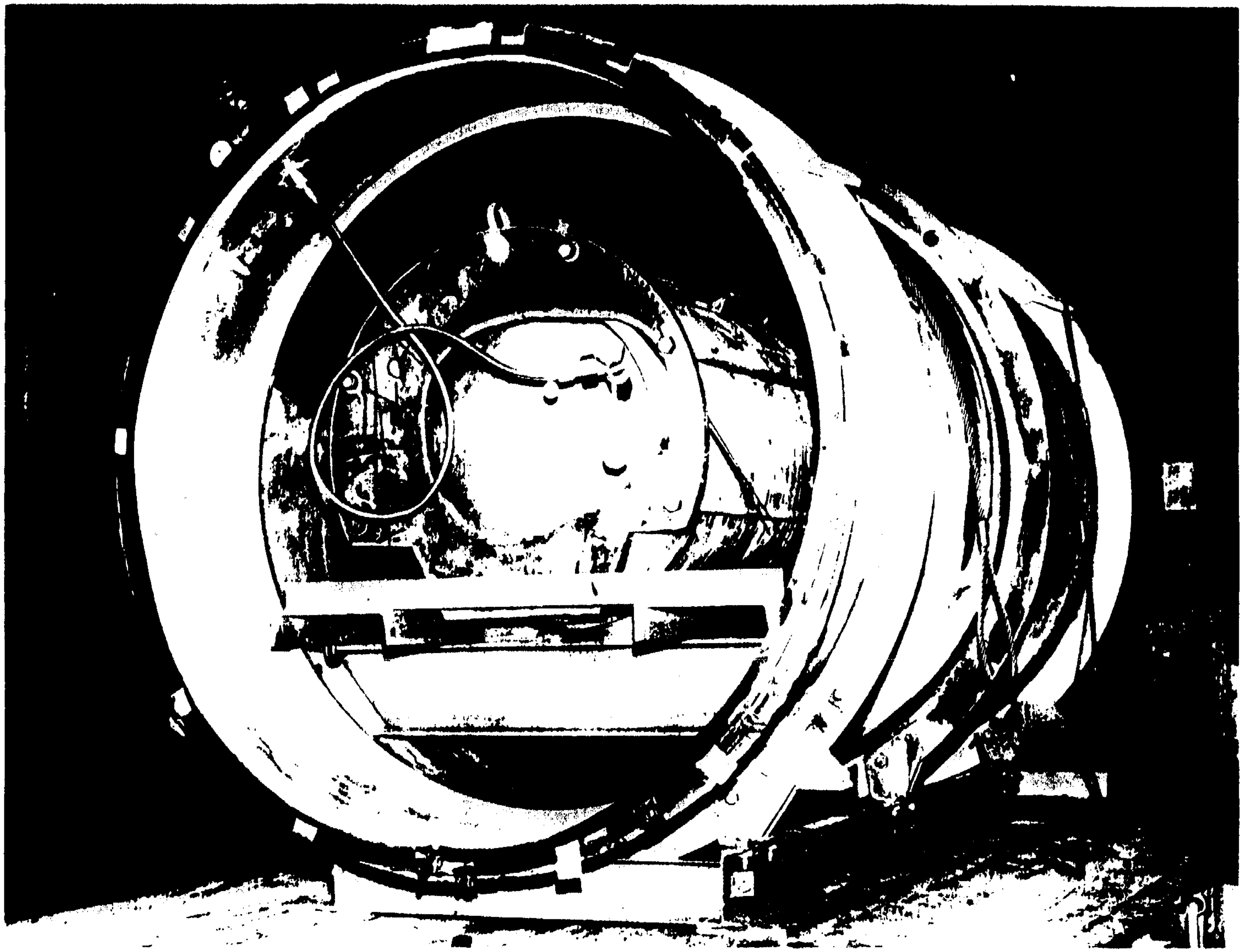

Figure 7

SECONDARY CONTAINMENT FOR DAMAGED CYLINDER

PHOTO NO. 75-3028

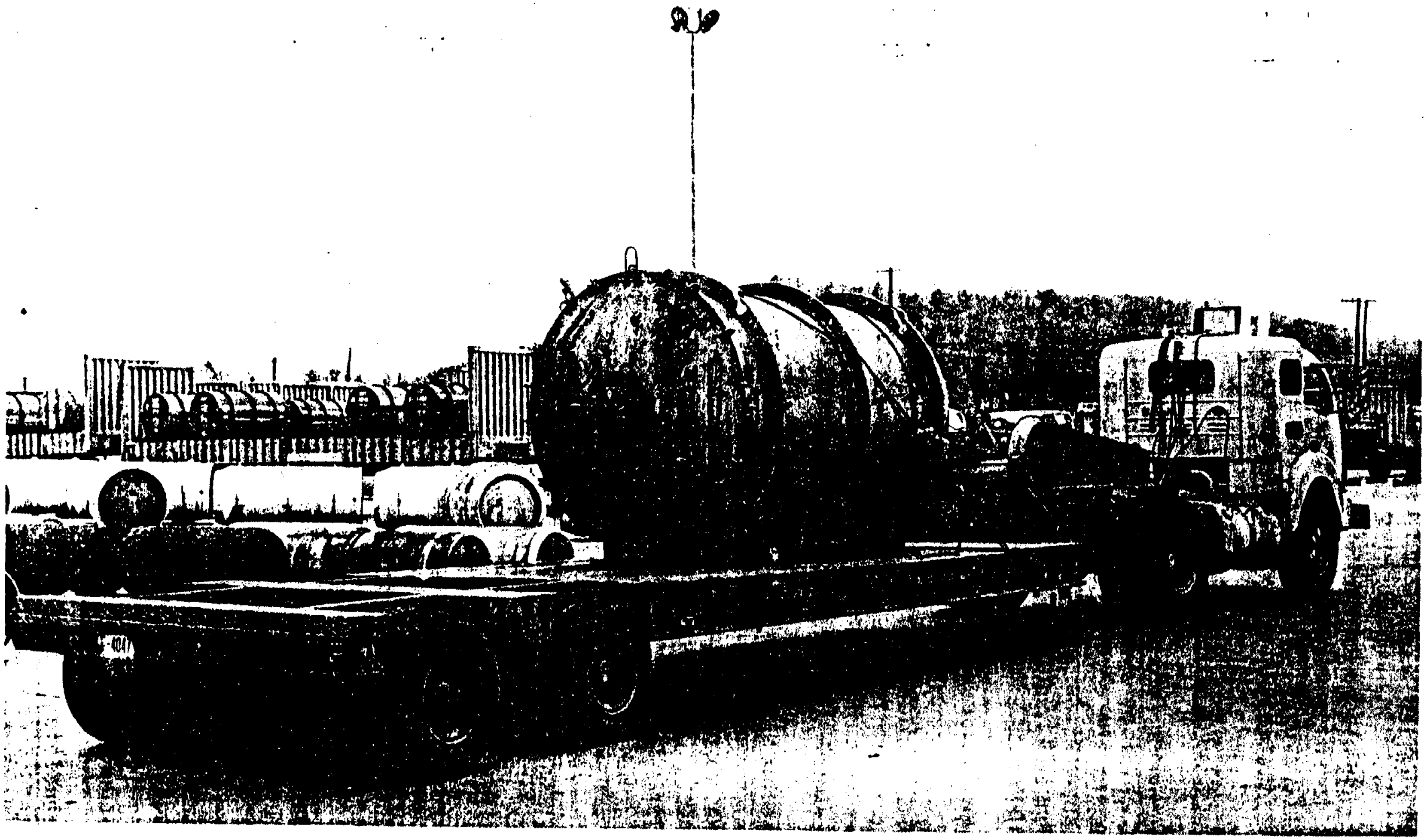

Figure 8
TRANSPORT OF CYLINDER ASSEMBLY TO CASCADE

Health Physics Impact

It was concluded by the Health Physics personnel that despite the potentials for high personnel exposure and facility contamination, actual exposure and surface contamination were quite limited. The Health Physics impact statement is given in Appendix D.

Metallurgical Evaluation Summary

30A Cylinder Design Evaluation: The 30A is a standard industrial Type D, 1-ton chlorine cylinder. It had been modified for UF_6 service early in the operation of the gaseous diffusion plants, and plant experience in its use through the intervening years has led to a stress analysis of the concave head, and later hydrostatic tests to failure and an evaluation of possible brittle failure characteristics.

Following the September 17 incident, the strain gage data from the early stress analysis and the results of the pressure tests to failure were reviewed. In addition, a plastic limit analysis of the cylinder head was made to obtain upper and lower bounds on its collapse pressure. Collectively this information indicated that the onset of head reversal could initiate as low as 600 psi with complete collapse occurring below 1300 psi.

Examination of Damaged Cylinder: Visual examination of the area near the crack in the cylinder wall revealed the presence of a buckle or kink in the wall extending about three-fourths of the total circumference of the cylinder. This kink suggested the presence of a crack in this area. Radiograph, dye penetrant, and ultrasonic examinations of this area confirmed the existence of the crack over at least 20 inches of circumference with additional indications being present but inconclusive.

Energy Estimates: To estimate the energy involved in the reaction within the cylinder, measurements were made on Cylinder N-8 at appropriate points, and measurements at identical locations were obtained on Cylinder N-1, apparently from the same manufacturing lot. The calculated volume change, and the plastic work involved, along with estimates of damage to the concrete pad, were the basis for energy estimates that indicated about 700,000 ft-lb of energy expended. Metallurgical considerations and details of the various calculations are given in Appendix E.

The characterizations of the explosive reaction in the Cylinder N-8 are based on a minimum of physical data regarding the volume change in the cylinder and on estimates of its mechanical properties. Final disposition of the cylinder will permit a measurement of its present volume, a detailed examination of the true extent of the crack at the plug end, and a full characterization of the static and dynamic mechanical properties, as well as chemistry and microstructure of the steel plates from the cylinder wall and heads.

Safe Handling of Damaged Cylinder: Even though ductile behavior was expected of the cylinder material under the temperature condition existing at the time of the explosive reaction, the known brittle behavior of some cylinder steels at ambient temperatures and the extent of the observed crack dictated the need for extreme caution in handling the cylinder to recover its contents. A fixture was designed and built which would assure that the cylinder end remained in place even in the event that the crack might propagate to completion. A clamp-on patch and a secondary containment vessel were also fabricated, both to minimize the possibility of further outleakage and to permit control of possible in-leakage during evacuation and recovery of the cylinder contents.

Analytical Investigation

The analysis of gas samples taken from the cylinder indicated that a reaction of a hydrocarbon and UF_6 had occurred. The chemistry of the incident and events that occurred just prior to and immediately after the UF_6 release are given in Appendix F.

Nuclear Criticality Safety Considerations

The cylinder contained UF_6 enriched to 3.28%. Under optimum conditions, i.e., spherical geometry and water reflection, about 2.5 kg of uranium-235 would be required for criticality; this is equivalent to about 113 kg (250 lb) of UF_6 in a volume of about 50 liters (10 gal) of water. An infinite length cylinder with about a 14-in. diameter or an infinite slab with a thickness of about 7.5 in. are other geometrical configurations which could be made critical at this assay under optimum conditions.

With the presence of nearby underground cable tunnels and a storm drain, combined with the rainy weather conditions, there was concern that the cylinder could completely fail, resulting in large quantities of released material collecting in these areas. Since no immediate action could be taken to improve the condition of the cylinder, precautions were taken to protect the cylinder from the weather and to prevent the uranium from entering the pits. The nuclear criticality safety considerations are presented in detail in Appendix G.

Safety Impact Statement

Three operators were directly involved in the movement of the cylinder. Mr. X was the operator of the forklift. Operators No. 1 and No. 2 were the operators to disengage the hooks from the cylinder.

No. 1 was at the valve end of the cylinder and was unhooking one end of the lifting fixture when the explosive reaction occurred. The hook was forced away from the cylinder striking his right hand which resulted in a sprain of his wrist. Material which was sprayed from the crack in the valve struck the employee on the leg. This resulted in a mild HF burn. No. 1 was treated at the plant medical center and returned to work on the same shift.

No. 2 was standing at the plug end of the cylinder (opposite end of valve) when the incident occurred. The rivets securing the metal name plate on the end of the cylinder were torn loose and the name plate was forcible ejected, striking the employee on the knee. The impact was sufficient to cause the employee to be knocked to the ground.

No. 2 was promptly transported to the medical center by ambulance, examined, X-rayed, and then taken to the hospital for further examination. The employee sustained a fracture of the knee-cap and partially severed tendon. The laceration was sutured and he was returned to the plant and placed on modified work.

At this time No. 2's injury was classified as a Recordable Injury. The employee continued to do modified work until October 10. Complication to the leg, as a result of the injury, was diagnosed as a blood clot and the case was changed from a Recordable Injury to a Disabling Injury. No. 2 has returned to work.

Standard Operating Procedures and Job Safety Analyses for cylinder handling and forklift operation were reviewed. They are adequate and were followed.

Industrial Hygiene Impact

At about 1:40 p.m. on September 17, word was received by way of radio in the safety office that an explosion had occurred at the Toll Enrichment Facility and UF_6 was being released to the atmosphere.

The Industrial Hygiene surveyor responded immediately to the area of the incident. Areas were sampled using Drager tubes for HF vapors. Upon arrival at 1:50 p.m. a sample was taken approximately 20 ft from the cylinder with negative results. A sample was taken within the building at 2:00 p.m. with no detectable amount of HF. The only sample taken which indicated any HF present in the air was taken in the dry ice fog, one foot above ground level, about 12 ft from the cylinder. The measured concentration of this sample was 10 ppm of HF. The threshold limit values for HF is 3 ppm for an 8-hour exposure and a 40-hour week.

Quality Assurance

The Quality Assurance Plans and associated procedures covering the areas of operation involved in this incident were reviewed. The results of this review follow:

Product Withdrawal: The following QA plan and operational procedures associated with product purity were reviewed.

QA Plan 6097 - Withdrawal of UF_6 Product

SOP 215.10 - Product Withdrawal Operations

SOP 325.7 - Product Withdrawal and Assay Control

The above plan and procedures establish purity control of the ORGDP product which is subsequently transferred into cylinders provided by the enriching services customer. The review and investigation revealed no abnormalities in either the product or the 14-ton "parent" cylinder used to supply the material for the customer owned cylinder involved in the incident. The QA plan and operational procedures were found to be adequate and were followed.

K-1423 Toll Enrichment Facility: The following QA plan and procedures were reviewed.

QA Plan 6104 - This plan itemizes the quality requirements for cylinders and product material.

SOP - Transfer of Liquid UF_6

SOP - Cold Pressure Check of UF_6 Cylinders

The above procedures describe the steps involved in filling a UF_6 cylinder for shipment. The QA plan and operational procedures were followed and found to be adequate. It is recommended, however, that a section describing special operating conditions should be added to the SOP on Transfer of Liquid UF_6 since at times it appears necesary to "burp" cylinders to facilitate proper filling later. The operation should be described and the precautions to be taken itemized.

K-1420 Cylinder Decontamination:

SOP 1410-5 Cylinder Decontamination - This procedure describes the techniques necessary to remove contaminants from within UF_6 cylinders including the removal of oils. The procedure was followed and found to be adequate. Investigation revealed that there is no QA plan per se for cylinder decontamination; one is being prepared.

K-1401 - Cylinder Valving: The following procedure was reviewed and evaluated:

MEP 300 - Repair and Valving of Model 30A UF_6 Cylinders - This procedure is used by personnel in the Valve Shop. It is quite detailed and calls for appropriate internal and external inspection by valve shop and inspection personnel. The one step that offers only a single contingency protection against malfunction states, "the incidence of a vacuum pump failure or inadvertent shutdown while pulling down a cylinder shall necessitate removal of the cylinder for degreasing and a cleanliness check." It is not possible to make an internal inspection of the cylinder following evacuation and prior to filling, as the configuration of the valve port precludes visual access to the cylinder interior. The evacuation system is not adequate in that it does not provide measures to prevent the introduction of oil into an evacuated cylinder in the event of a failure in the system. Investigation revealed that no QA plan exists for the valving and repair operation. One is being prepared and the vacuum system is being modified to prevent future occurrence of a similar incident.

INVESTIGATIVE FINDINGS AND CONCLUSIONS

1. Analyses of reaction product gas removed from Cylinder N-8 after containment established that an explosive reaction involving a hydrocarbon and UF_6 was the cause of the incident.

2. It was determined that oil was probably introduced into Cylinder N-8 as a result of a motor failure on a vacuum pump used to evacuate the cylinder.

3. The two operators who were adjacent to the cylinder when the explosive reaction occurred sustained injuries and received medical attention. One of these employees returned to normal work after treatment. The second employee sustained a fracture of the knee-cap and a partially severed tendon and was initially placed on modified work. Because of complications which developed later this became a disabling injury.

4. The QA plans for UF_6 product withdrawal and the K-1423 Toll Enrichment Facility are considered adequate.

5. Cylinder decontamination procedures are considered adequate; however, a QA plan has not been provided.

6. The valve shop system for cylinder evacuation was not considered adequate. A QA plan for the valving operation in the shop is needed.

7. Overall plant operations were not affected by the incident. Normal operations at the Toll Enrichment Facility were resumed at the beginning of the next scheduled shift.

8. The local Emergency Squad and the Plant Emergency Squad did an effective job in containing the release. Fire and Guard Department personnel also performed in a very effective manner.

9. Cylinder weighings indicate a release of approximately 18 lb of enriched UF_6 (3.2851% uranium).

10. The estimated cost attributable to the incident amounts to $5,362.81.

11. Fifty-three employees, including those checked as a precautionary measure, submitted urine samples for analysis. The highest personnel exposure corresponded to an intake for 1 quarter of 17% of the ERDA Radiation Standard for soluble airborne uranium. Over half of these 53 employees indicated uranium intake of less than 1% of the ERDA standard.

12. Alpha radiation surveys of the area following the incident indicated that contamination above 500 counts/min/100 cm^2 was limited to an area within a 20-ft radius of the damaged cylinder.

13. The environmental impact of the release was insignificant in that no significant quantities of uranium, and fluorides were detected beyond the plant boundary.

14. Area radiation surveys and analysis of reaction products from the damaged cylinder indicated the absence of fission products which would have been expected from a nuclear excursion.

15. Based on the cylinder volume expansion as measured on Cylinder N-8, the total energy involved in the reaction can be assumed to be about 230 kcal.

16. Using the nature of the gaseous products generated, and the nature and quantity of reduced uranium fluorides produced (304 lb), it is estimated that the quantity of hydrocarbon involved was between 1.0 and 1.6 liters. The minimum quantity actually required to generate the energy required is only about 30 cc which indicates a low order of efficiency for the reaction. The reaction appears to be one which requires the presence of liquid UF_6 and time to generate more reactive fluorohydrocarbon residues before the rate of reaction becomes fast enough to produce the explosion.

17. There were insufficient impurities in the 14-ton "parent" cylinder to have caused the incident.

18. Analysis of the cold trap used for evacuation of Cylinder N-8 burping operation showed no gaseous reaction products which could have been produced during the cylinder filling operation.

19. The incident must be viewed as a "near miss" of a more serious incident in that the potential existed for complete separation of the cylinder head and the more serious consequences therefrom.

20. Investigation confirmed in most areas that the QA plans and related operating procedures and Job Safety Analyses existed, were adequate, and were followed. Areas were identified where plans are needed. Division management has established a schedule to be pursued with systematic follow-up to ensure that necessary improvements are made.

RECOMMENDATIONS

1. To preclude the introduction of reactive contaminants into cylinders used in UF_6 service, it is recommended that the evacuation of these cylinders be accomplished by use of equipment such as air ejectors. If other equipment such as oil lubricated pumps or compressors is used for evacuation or pressuring of cylinders, a safety analysis should be made to assure design of equipment and its operation precludes the introduction of reactive contaminants into the cylinder. Design considerations should include traps, filters, and check valves.

2. QA plans will be prepared for cylinder decontamination and the cylinder valve installation operation in the valve shop.

3. ERDA should make available to Nuclear Industry licensees and vendors involved in UF_6 handling the conclusions and recommendations of this incident investigation.

   ERDA should also formally notify all foreign and domestic owners of UF_6 cylinders of their responsibility to assure that all cylinders and valves comply with ANSI-N14.1 specifications and that they provide written certification that these specifications have been met prior to delivery of the cylinders to the gaseous diffusion plants. An approved vendor QA plan should be required. It is further recommended that the owners of UF_6 cylinders be notified of their responsibilities in regard to the 5-yr retest and cleanliness of these cylinders as specified in ANSI-N14.1.

4. The report of this incident should be reviewed by the Three-Plant UF_6 Cylinder Handling Committee to determine whether existing committee recommendations are adequate to prevent recurrence of such an incident.

5. A complete examination of Cylinder N-8 should be conducted. This examination should include analysis of residues in the cylinder, measurement of the current volume, a detailed examination of the extent of the crack in the cylinder, and a full characterization of the properties of the cylinder steel. The results of this examination should be formally reported.

6. Procedures should be expanded to include those infrequent operations at Toll Enrichment Facility such as cylinder burping.

7. Additional sampling capabilities should be installed to improve the quantification of offsite concentrations of uranium and fluorides resulting from accidental UF_6 releases.

SIGNATURES OF INVESTIGATING COMMITTEE

A. J. Legeay, Chairman

H. R. Dyer

L. W. Anderson

J. Dykstra

J. W. Arendt

E. J. Kimmerly

J. C. Bailey

for M. E. Mitchell

E. J. Barber

R. L. Newton

J. C. Barton

J. W. Pickel

T. B. Bomar

K. T. Ziehlke

O. L. Calvert

APPENDIX A

VALVE SHOP EVACUATION PUMP TEST

APPENDIX A

VALVE SHOP EVACUATION PUMP TEST

J. Dykstra

Description

A number of tests were conducted on October 1, 1975 in the Maintenance Shop where the cylinders are fitted with valves and evacuated in an effort to understand how oil might be unintentionally and undetectably introduced into a cylinder. Failure of the vacuum pump was simulated to demonstrate that oil could be transferred from the vacuum pump into the cylinder if operation was interrupted during an evacuation operation.

A four-inch diameter pipe trap was connected to an evacuation hose between the Kinney vacuum pump and a 30A UF_6 cylinder. The vacuum pump was operated until the cylinder was evacuated to ~27-inch vacuum. The vacuum pump was then shut down to determine the quantity of oil transferred from the Kinney pump as the pressure in the evacuated cylinder equalized with the pump. This oil was collected in a 4-inch diameter trap which had been tare weighed. The trap was removed for final weighing after the pressures had equalized.

Observations

Oil Collector Weight:

Initial: 12,796 g. Final: 14,109 g. Net gain: 1,313 g.

Time	Pump Suction Pressure	Remarks
10:31 a.m.	Atmospheric	Pump started.
10:32 a.m.	---	Valve opened to system.
10:37 a.m.	27-inch vacuum	Pump shut down.
10:40 a.m.	25-inch vacuum	Oil back flow started.
10:47 a.m.	Atmospheric	Pressure equalized.

Conclusion

This test verified that stoppage of the pump during the evacuation of a cylinder could result in the transfer of sufficient oil to the evacuated cylinder to have produced the observed result as indicated by the calculations of the minimum quantity required in Appendix F.

Other tests simulating pump failure resulted in transfer of quantities ranging from 250 cc to more than 2000 cc of oil. Similar results were obtained by closing the evacuation manifold valve when the cylinder pressure was 20-inch vacuum.

Studies of the effects of pump oil on the surgical rubber hose used to connect the vacuum pump to the cylinder indicated that a film of oil on the surface of the hose diffused into the wall leaving a dry surface. This indicates that unless an oil flow into the cylinder was observed when it occurred, examination of the hose at a later time would not necessarily give any indication of oil having passed through it.

APPENDIX B

PLANT EMERGENCY DIRECTOR'S STATEMENT

APPENDIX B

PLANT EMERGENCY DIRECTOR'S STATEMENT

R. L. Newton

Action at Time of Incident

A supervisor reported a UF_6 explosion incident to the Shift Superintendent at Station 9 at 1:30 p.m. and requested an ambulance and assistance with a leaking cylinder. The request was transmitted to the Fire Department and the Plant Emergency Squad was paged on all three radio channels.

Station 9 attempted to contact Health Physics personnel, contacted Plant Emergency Director and answered dozens of phone calls, some of which were of a curiosity nature.

When the Fire Department personnel arrived, an injured employee was on the ground. He was loaded into the ambulance and taken to the Medical Center and to the hospital.

Two Emergency Squad members were dispatched to pick up dry ice. Another Emergency Squad member delivered the Emergency Truck to the scene. One Fireman stood by with the fire pumper.

The Toll Enrichment Facility local Emergency Squad did an outstanding job in containing the release which was controlled within 10 minutes and stopped within 20 minutes. A 50-pound CO_2 fire extinguisher was used, wet towels were placed over the leaks, and a controlled stream of water from a 3/4-inch rubber hose was directed on the middle of the cylinder to avoid the introduction of water to the ruptured points on the cylinder. The water flow was discontinued when the dry ice was applied. Emergency Squad members assisted with the application of dry ice to the damaged cylinder and set up air samplers. Guards established road blocks to control traffic in the area. Two firemen and one Emergency Squad member were dressed in Scott Air Packs and could have been used to assist in containing the release had it been necessary.

Action at Time of Packaging, Weighing, and Transporting the Damaged Cylinder

On September 26, 1975, plans were formulated for placing the cylinder in a secondary containment vessel, weighing the package, and transporting it to the feed point for evacuation. Minor gas releases occurred when the valve was replaced. These were contained with vacuum collectors. No emergency conditions developed during the entire operation. The following precautions were taken to nullify an emergency if one had occurred.

1. Road blocks were set up to restrict traffic past the Toll Enrichment Facility and into the immediate area.

2. A fire truck, the Emergency Truck, and the Criticality Control Unit with borated water pumper were positioned upwind of the activity area.

3. Two Emergency Squad members were dressed out in Scott Air Packs, with other protective equipment, throughout the operation.

4. The Emergency Power Plant, equipped with flood lights, was started and placed in an upwind position.

5. Escorts were provided during the transport to the feed point.

In summary, a potentially serious situation existed and it was handled successfully without UF_6 exposure or property damage. The weather conditions were ideal--raining and little or no wind. A clear day with 10 mph winds could present a much more severe problem.

The local Emergency Squad did an exceptionally good job in containing the release. The Plant Emergency Squad performed all tasks assigned to them. The Fire and Guard Department personnel performed in a professional manner.

APPENDIX C

ENVIRONMENTAL IMPACT STATEMENT

APPENDIX C
ENVIRONMENTAL IMPACT STATEMENT
M. E. Mitchell

Response to the Incident

The ORGDP Environmental Management Group learned of the incident at approximately 1:55 p.m. Members of the Environmental Management Team went to the site, where they were informed that a 2.5-ton cylinder of product UF_6 had ruptured while being placed in a cooling position just west of the K-1423 facility. Although no evidence of a release existed at that time (approximately 2:00 p.m.), it was evident, from conversations with eyewitnesses, that a finite quantity of material had been released. Therefore, laboratory and utility personnel were contacted immediately and instructed to actuate all ambient air particulate samplers as well as the water sampler servicing the drainage basin around K-1423. All of the air samplers were operating by about 2:25 p.m. and continued to operate for the following 22.5 hours. The water sampler, which is located at the K-1700 discharge, was inoperable; however, grab samples were collected every two hours from about 2:30 p.m. on the day of the release until 2:30 p.m. the following day.

Qualitative Description of the Release

According to the accounts of eyewitnesses, the cylinder rupture was followed by a steady, high-velocity stream of gas emanating from the west end of the cylinder; this stream continued for about 10 minutes. Due to the orientation of the rupture (or crack) in the cylinder, this gaseous stream was projected in a horizontal southwesterly direction. As the UF_6 contacted the moist air, it reacted with the water to produce gaseous HF and particulate UO_2F_2. The visible plume resulting from the release, which was apparently HF and condensed water droplets, was also observed to be moving in a southwesterly direction. All descriptions of this plume indicate that it was visible only to the southwest extremity of the K-1423 area and that its maximum height was about 25 feet above the ground. The bottom of the plume was apparently 3 to 4 feet above the ground. Since the wind was about 5 mph toward the west, it is probable that the initial momentum of the release coupled with the effects of the K-25 and K-1423 buildings would have caused the plume to continue to move in a southwesterly direction.

No material was observed on the concrete surface surrounding the K-1423 facility.

The Dissipation of the Released Material

As indicated previously, the total release was determined to be less than 18 pounds of material. Assuming all the material was released as UF_6 this would have resulted in a release of less than 15.8 pounds of UO_2F_2 (less than 12.2 pounds of total U) and less than 4.1 pounds of HF. Since a light rain was falling at the time of the release, it is probable that all of this material was deposited within the general area of the K-1423 building. However, in order to provide a thorough assessment of the maximum as well as the probable environmental effects of this release, all possible mechanisms of dissipation have been analyzed. These mechanisms are described below along with the environmental concentrations and subsequent impacts they could have incurred.

A. Atmospheric Dispersion

The gaseous plume observed during the release should have consisted primarily of HF and condensed water, with only a small quantity of entrained UO_2F_2. As mentioned previously, this plume apparently moved in a south, southwesterly direction, where the nearest perimeter fence is about 2600 feet (790 meters) from the site of the release. The atmospheric concentration of uranium at this location, as well as at three other locations around the plant, was determined to be slightly less than the normal concentration (0.016 ug/m^3), during the period between 2:25 p.m. on September 17, 1975 and 12:55 p.m. on September 18, 1975. These data were collected by four high-volume air samplers. However, due to the time lapse between the release and the startup of the air samplers (about 50 to 55 minutes), it is possible that the plume (moving at about 5 mph) passed over the perimeter fence without being sampled. Thus, the data collected, although negative in nature, do not alone conclusively prove that no uranium or HF escaped the plant boundary.

As a second means of assessing the flight of the plume, a mathematical model[1] incorporating the meteorological conditions of that day was used to calculate maximum possible uranium and HF concentrations at the site boundary. Based on the accounts of eyewitnesses, a release duration of 10 minutes was incorporated into the model.

This resulted in a release rate of about 9.3 grams of uranium per second and 3.1 grams of HF per second. Assuming that all of the uranium was entrained within the plume, the maximum concentration of uranium at the perimeter fence 2600 feet away would have been about 5.5 mg/m^3. The same assumption for the HF would have resulted in a maximum perimeter fence concentration of 1.8 mg/m^3 of HF.

A person standing at the perimeter fence in the middle of the plume for ten minutes would then have received a radiation dose equivalent to approximately 2% of the ERDA weekly limit for alpha activity. The 5.5 mg/m^3 of total uranium, when averaged over a period of one week, is approximately 2.4% of the ERDA chemical toxicity limit.

The same person would have detected the odor of the HF but would not have experienced any discomfort since, according to the National Academy of Sciences,[2] the threshold for such discomfort is 25 mg/m^3. A concentration of 2.0 mg/m^3 is the recommended 8-hour exposure threshold limit.[2]

No data are available to predict the effects of such small short-term releases of uranium and HF on terrestrial vegetation. Therefore, considering (1) that the rain on the day of the release probably "scrubbed" or "scavenged" the majority of the uranium and HF from the atmosphere and deposited it on the ground in the general area of the release; and (2) that the maximum concentrations of HF and uranium that could have occurred at the perimeter fence, if all material were dispersed that far, would not have been detrimental for the short time they could have existed; the net effect of the atmospheric dissipation of the HF and uranium would, at the worst, have been minimal.

B. Surface Stream Dissipation

The entire area around the K-1423 facility is located within the K-1700 drainage basin. The effluent from this basin, which includes the discharge from the K-1407 holding pond, enters Poplar Creek at a point about 500 feet downstream of Blair Bridge. The flow rate of this effluent averages about 820 gpm. The flow rate during the 24-hour period following the release ranged between 1160 and 1880 gpm, with the average being 1516 gpm; this higher than usual flow was due to excessive rainfall.

Since it is highly probable that the majority of the uranium and HF lost from the ruptured cylinder was deposited within this drainage basin, it is also highly probable that any material contained in surface runoff would have been discharged through the K-1700 discharge point. A total of 13 samples (one every two hours) was collected during the 24-hour period beginning at 2:30 p.m. on the day of the release and lasting through 2:30 p.m. on the following day. The uranium concentrations in these samples ranged from 0.049 mg/l to 0.239 mg/l, with the average being 0.102 mg/l. The average quantity of uranium discharged during this period was 33.6 g/hr. The assay of this material ranged from 0.89 to 1.08% U-235. The fluoride concentrations in the 13 samples varied between 1.2 and 2.4 mg/l, with the average being 1.8 mg/l. The quantity of fluorides discharged was about 623 g/hr.

In assessing the data collected during the 24 hours following the release, several sets of historical data have been used for comparison, including one sample collected the day before the release, September 16, 1975. These data indicate that the normal uranium discharge through K-1700 is actually slightly higher (43.1 g/hr. vs. 33.6 g/hr.) than that recorded on the day following the incident. The assay of the K-1700 uranium discharge usually ranges from 1.1 to 1.3% U-235. It is thus apparent that no detectable quantity of the uranium lost from the incident was discharged through the K-1700 effluent.

The fluoride discharges from K-1700 during the 24 hours following the release were about 3.5 times higher than normal. However, since the K-1420 facility was decontaminating sodium fluoride traps that same day, with the waste stream being discharged through the K-1700 point, it can be stated positively that all of this abnormal fluoride discharge was not a result of the release from the ruptured cylinder. Unfortunately, no means exist for determining the contributions of this release. It should be noted that the fluoride discharges from K-1700 are not now restricted by the Environmental Protection Agency, and thus, no regulation was violated. Even when these discharges do become restricted (January 1, 1977), the limit will be 20 mg/l of F^- in the K-1407-B effluent, which extrapolates to about 16 to 17 mg/l at the K-1700 point. The F^- concentration following the incident was thus only about 11% of the future EPA limit.

After determining that no detectable quantity of the uranium lost from the cylinder had been discharged to Poplar Creek or dissipated off-site through the atmosphere, an effort was made to determine the actual disposition of the material. The two locations having the greatest opportunity for collecting the uranium were the electrical duct underneath the K-1423 parking lot and the sediments of the K-1700 pond. A sample of the water in the duct revealed a total uranium concentration of about 4.9 mg/l, with a 3.22% U-235 content. It is thus apparent that some portion of the released material was contained within the electrical duct. However, since no estimate of the volume of water in the duct is available, no estimate of the total quantity of uranium contained is available. The silt in the bottom of the K-1700 pond was found to contain about 2080 ppm by weight of total uranium with an assay of about 1.4% U-235. Since these values are extremely close to those normally found in the K-1700 and K-1407-B ponds, it was concluded that no significant quantity of the lost product material was found in the K-1700 pond sediments.

References for Environmental Impacts Section

[1] Smith, Maynard, "Recommended Guide For The Prediction of the Dispersion of Airborne Effluents," The American Society of Mechanical Engineers, New York, N. Y., 1968.

[2] National Academy of Sciences, "Biological Effects of Atmospheric Pollutants: Fluorides," U. S. Government Printing Office, Washington, D.C., 1971.

APPENDIX D

HEALTH PHYSICS IMPACT STATEMENT

APPENDIX D

HEALTH PHYSICS IMPACT STATEMENT

J. C. Bailey

Personnel Exposures and Facility Contamination

Personnel Exposures: The highest personnel exposure* as indicated by urinalysis data corresponded to an intake of uranium equal to that which would be received by an employee breathing air for one quarter at 17% of the ERDA Radiation Standard for soluble airborne uranium. The short-term intake of this amount of material would not result in a concentration of uranium in any body organ in excess of applicable guide values, even temporarily.

As a precautionary measure, all persons who were recognizably subject to possible uranium inhalation submitted urine samples for analysis. Sampling involved a total of 53 persons. Indicated uranium intake for over half of these corresponded to exposure for a quarter to less than 1% of the ERDA Radiation Standard for airborne soluble uranium. The distribution of indicated exposures is shown graphically in Figure 1.

Two air samples covering the same time interval were taken by emergency personnel for the period from 40 minutes to one hour ten minutes after the initial release. One taken north of the K-1413 building and closely downwind from the cylinder indicated an airborne concentration of uranium equal to 3% of the ERDA Radiation Standard for industrial exposure to soluble uranium as related to uranium mass concentration, or to 2% of the standard as related to alpha activity. The second sample, taken northeast of the K-1413 building, showed no detectable alpha activity.

In view of the time relationship to the initial release, these data are not considered indicative of exposure potentials for the incident. As noted previously, primary reliance for evaluating actual uranium intake is placed on urinalysis data.

Facility Contamination: Alpha radiation surveys of the area on the day following the incident and after surfaces were dry enough to permit alpha monitoring indicated that contamination above 500 counts/min/100 cm^2 was limited to an area within a radius of about 20 ft from the cylinder.

Adjacent roadways, parking areas, and surfaces adjacent to the K-1423 building showed no detectable contamination.

A plot of contamination levels in the immediate vicinity of the cylinder prior to removal of the cylinder and cleanup of the area is shown in Figure 2.

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*Mr. X , forklift operator.

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(U)

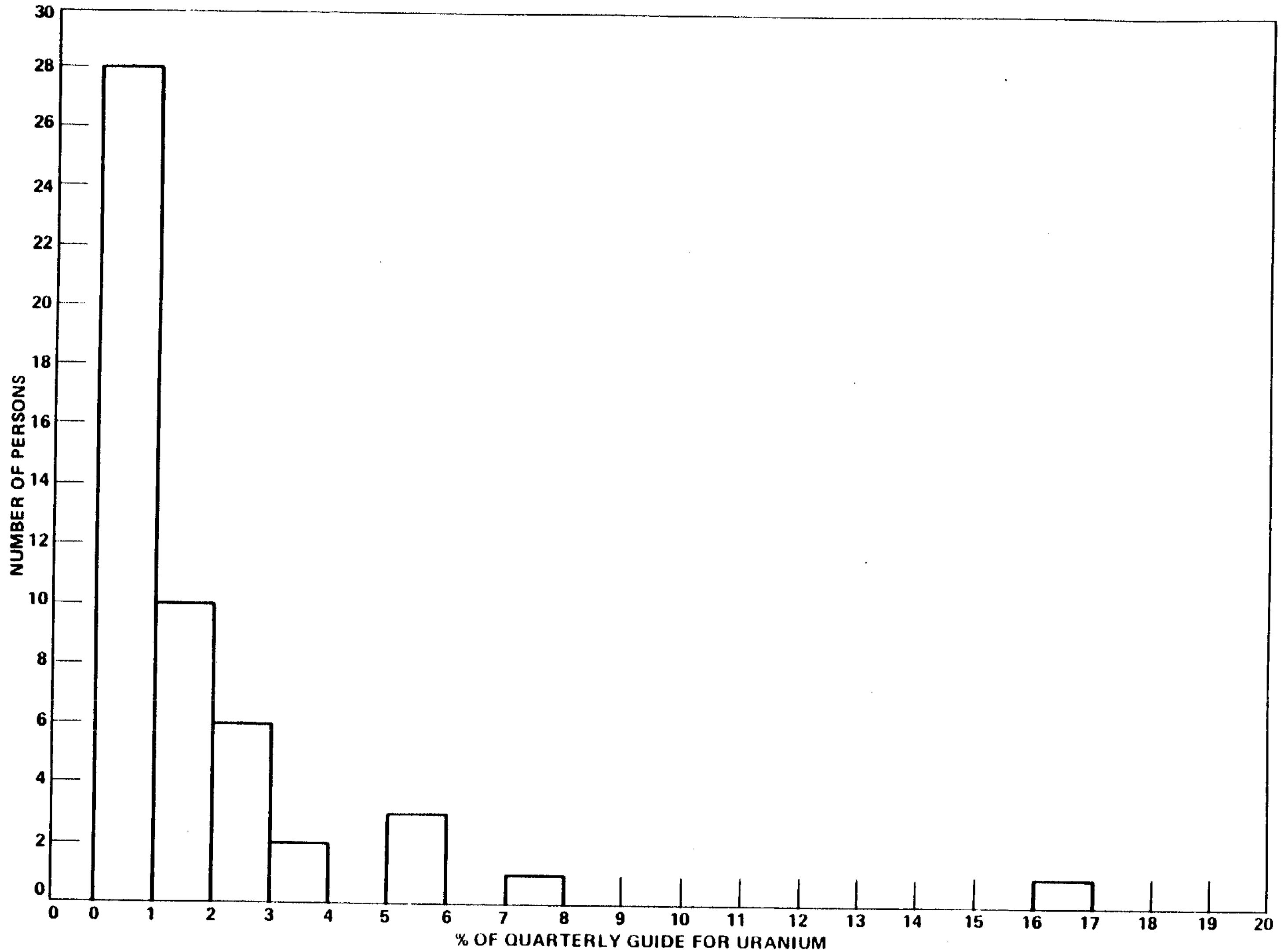

Figure 1

DISTRIBUTION OF INDICATED EXPOSURES
TOTAL PERSONS - 53

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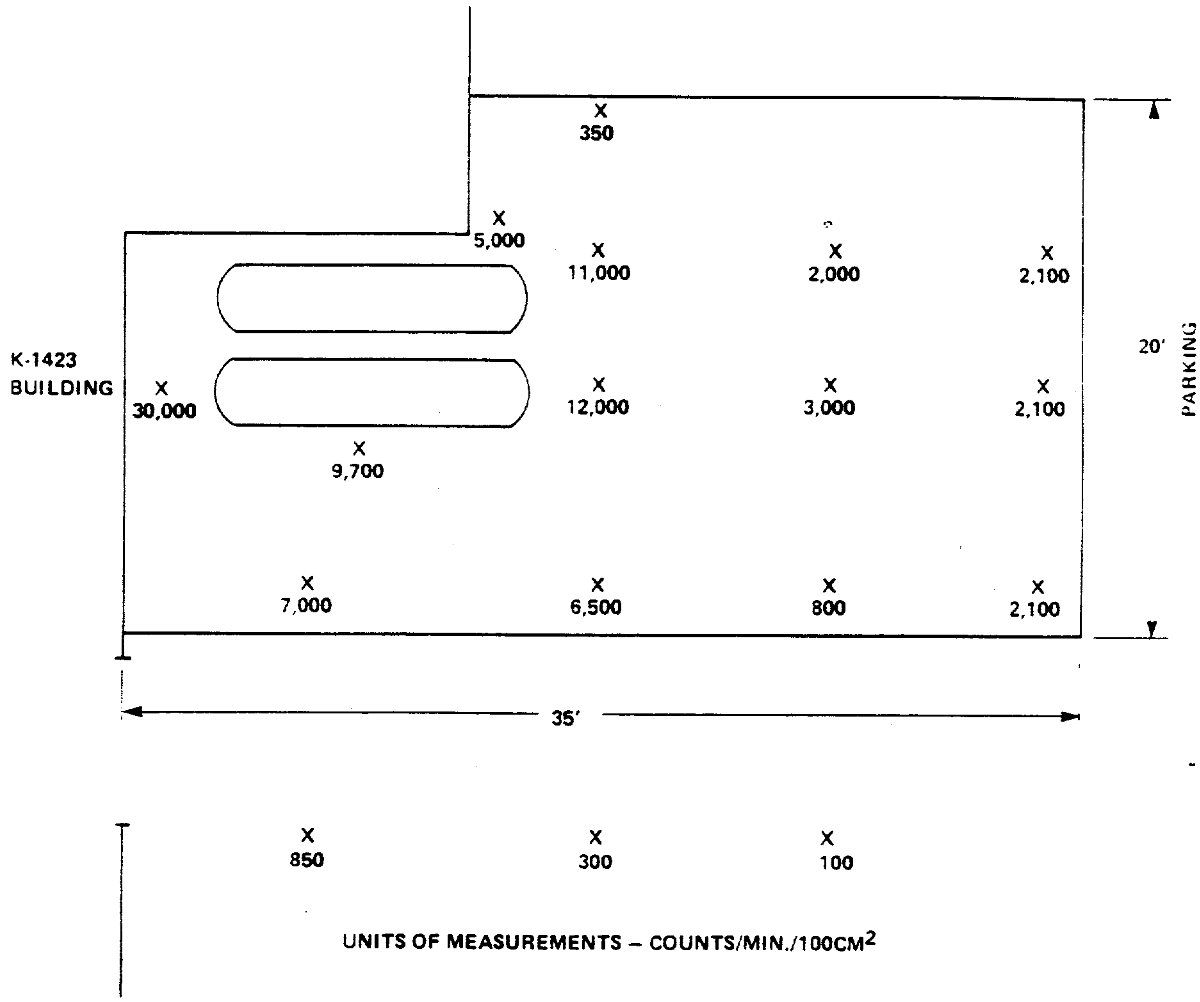
(U)



CONTAMINATION LEVELS PRIOR TO CLEANUP K-1423

FIGURE 2

In summary, actual exposures and surface contamination were quite limited despite the potentials of high personnel exposure and facility contamination associated with the release of uranium materials from the cylinder in this incident. The occurrence of heavy rain during the period immediately after the release would probably have carried much of the uranium away in the run-off water, thus limiting surface contamination found in subsequent surveys.

Investigation of Nuclear Excursion Possibility

The possibility that the rupture of the cylinder may have been due to a critical nuclear reaction within the cylinder was examined.

Radiation Alarms: The fact that neither the radiation alarm at K-1423 itself nor the one at the nearby K-1413 building indicated a nuclear excursion gave immediate assurance that the incident was not nuclear in nature. These devices are activated by the accumulation of a dose of less than 12 mr gamma within a 30-second period or an equivalent response to neutrons, and are thus not dependent on a sustained radiation dose rate for activation. The detector chambers are coated with boron to assure neutron sensitivity.

Prior to their adoption for use at ORGDP the instruments were tested extensively at the Los Alamos Godiva fast-pulse reactor and their satisfactory response to a fast nuclear excursion was demonstrated.

It should be recognized that in the event of a critical reaction, the power level of the reaction will increase exponentially until sufficient energy is released to alter the critical assembly in some way and thereby limit or terminate the initial reaction. The occurrence of a very low level fast excursion in the absence of some external control mechanism is thus a physical impossibility. It is thus assured that any fast-pulse nuclear excursion would be of sufficient magnitude to activate nearby alarms. In the present case, the physical bulging of the cylinder ends gave direct additional evidence that a rather large amount of energy release was involved. The magnitude of a critical reaction required to produce these effects and estimates of radiation levels which would result from such a reaction are taken up in a later part of this section.

Despite the immediate assurance given by the radiation alarm devices that the occurrence was not nuclear in nature, additional data related to this possibility were accumulated and are discussed below.

Radiation Surveys: Detailed radiation surveys carried out on the day following the incident failed to indicate any radiation not due to the uranium material and the associated uranium daughter products.

Because of the vary large number of different isotopes which result from nuclear fission and which exhibit half-space lives varying from fractions of a second to many years, gamma radiation levels associated with a nuclear excursion decrease* approximately as (time following fission)$^{-1.2}$. Thus the immediate high levels of gamma radiation decay rapidly but residual gamma levels are sustained for very long periods of time.

It may thus be anticipated that gamma radiation from an assembly that has exhibited a fast-pulse reaction, especially one demonstrated by the condition of the cylinder to be rather large, would be detectably elevated for a period of months and perhaps even for years.

The absence of any detectable elevation in gamma levels on the day following the excursion was considered to further rule out the possibility that the reaction was nuclear in nature.

Personnel Monitoring Results: The film badges of four employees closely associated with the incident were examined. Upper limits for gamma exposure that might have been due to the incident were established from the gamma-monitoring film of the badges. Gross gamma readings varied from 200 mr to less than 50 mr.

It is noted that the incident occurred near the close of a 3-month use cycle for the films, and the film readings represented essentially the total quarterly dose for the persons involved. The measured low levels of exposure are not indicative of exposure to a critical reaction.

The bare gold foils and sulfur pellets, which are incorporated in the badges for emergency criticality monitoring, were examined. Neither of these detectors in any badge showed any activation. At the time of measurement 21% of the initial gold activity and 75% of the P-32 activity induced in the sulfur by neutron irradiation would have been present.

Magnitude of Energy-Equivalent Nuclear Excursion: The calculated energy release of about 230 kcal reported elsewhere in this report corresponds to the energy release from 3.0×10^{16} fissions.

An order-of-magnitude estimate of radiation exposures which would be associated with such a reaction occurring in a low moderation assembly may be obtained by direct comparison with the bare Health Physics Research Reactor at ORNL. A pulse of 3.0×10^{16} fissions in this assembly delivers a dose of 129 rad at a distance of 3 meters from the reactor. For persons immediately adjacent to the cylinder at the time of occurrence it therefore is apparent that exposures would have been on the order of at least 100 rad. Such an excursion with the attendant radiation exposures would be readily apparent from any of the detection mechanisms discussed above.

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*Glasstone, Samuel (editor), The Effects of Nuclear Weapons, Superintendent of Documents, Washington, D.C., June 1957.

It is evident that the reaction in the cylinder was not nuclear in nature.

In summary, the impact of this incident on health physics aspects of plant operations was relatively insignificant.

APPENDIX E

METALLURGICAL EVALUATION

APPENDIX E

METALLURGICAL EVALUATION

K. T. Ziehlke

Metallurgical Considerations

The 30A cylinders, designed for industrial chlorine service, were made from a class of mild steel plate covered by ASTM A285. This specification (the 1967 version applicable to N-8) required compliance with chemistry (Grade A: C.15 max, Mn.80 max, P. 0.035 max, S 0.04 max) and mechanical properties (Grade A firebox quality, UTS 45 - 55 ksi, YS 24 ksi min, 27% elong. in 8" min., and a bend test). No impact properties were required, although some cylinder handling and emergency procedures are carried out at low temperatures.

The reaction within the cylinder took place when the contents and the steel were at an approximate temperature of 190 - 200°F. At this temperature, the steel is expected to behave in a ductile manner; defects can propagate in brittle fashion only under conditions of restraint not likely to be encountered in the existing cylinder geometry. Any cracks developed during the reaction thus probably stopped growing immediately after the peak of the reaction pressure pulse. Subsequent cooling of the cylinder and its contents, and packing dry ice around the cylinder, would have subjected the structure to only moderate thermal stresses and thus the likelihood of additional crack extension was low.

In handling the cylinder to initiate recovery operations, two additional considerations were presented. Impact loading of even a relatively mild level could cause crack extension if the cylinder steel had a ductile - brittle transition temperature in the ambient temperature range[1], and it could also stimulate further explosive reactions within the cylinder. Either occurrence could easily result in completing the rupture at the plug end of the cylinder. A restraining fixture was affixed to the cylinder to minimize loss of the contents in the event of such a complete rupture. Attachment of the fixture and torquing of the tie bolts could have resulted in minor extension of the existing cracks through the flexing action or due to axial compression of the cylinder, but this was considered less likely than crack extension due to the anticipated stresses incurred during lifting or from pressure stress during the subsequent recovery or from thermal stress incurred during the initial cool-down following the incident.

Stresses of the same order of magnitude may result from evacuation of the cylinder. Here again, it is not considered likely that extensive crack growth can occur under relatively static and low loading conditions. Since it was planned to move the cylinder into a secondary containment vessel, the only apparent consequence of crack extension within the restraining fixture is increased inleakage of the buffer atmosphere.

Cylinder Integrity Considerations

Examination of the cylinder in place at K-1423 following the incident showed complete head reversal at both ends, a crack and leak in the valve body, a crack and leak in the cylinder wall at the plug end, and a relatively sharp reversal of curvature (a kink) in a circumferential direction extending on both sides of the crack (See Figure 1). The concave heads are forge welded to the cylindrical shell for a distance of about 2" from the cylinder end; the location of the kink corresponded to the end of the weld zones and suggested a partially through crack extending at least 3/4 of the circumference of the cylinder. Pressure tests to failure on two cylinders of this type[1] gave ruptures at this same location with similar head reversals. The characteristic failure mode was gradual head reversal beginning at 600 - 1000[2] psi and increasing progressively to rupture at 1050 - 1250 psi. Post-test sampling of the cylinder materials for the Charpy impact test showed a possibility of ductile-to-brittle transition temperatures in the 60 - 80°F range. The existence of a crack of greater-than-critical size and the ambient temperature in the transition temperature range dictated the need for extreme caution in handling the cylinder in order to avoid the possibility of carrying the partial failure to completion and actually separating the end from the cylinder.

Means for lifting the cylinder without additionally loading the cracked end were developed which allowed prior attachment of a clamping fixture to restrain the cracked end. The tie bar fixture was designed by Maintenance Engineering to keep the cracked end in place under a compressive axial load, and could support a substantial internal pressure if needed (about 200 psi at the yield strength of the tie bars).

While the lifting and restraining fixtures and a secondary containment vessel were being prepared, measurements of the actual extent of the plug-end crack were made. Six radiographs, covering about 80% of the flange area circumference, were made with an Iridium 192 source*. These radiographs, some of which are illustrated in Figures 3 through 6, showed features that were interpreted as cracks covering approximately 20 inches in length, from an area slightly below the through crack upward past the top of the cylinder (See Figure 2). Additional verification of the existence of cracks in this and other areas was obtained with an ultrasonic shear wave technique**. The ultrasonic scan showed positive indications (cracks or other discontinuities) in the areas where the radiographs showed cracks, and also gave intermittent signals in some of the kink areas where the radiographs were inconclusive. The quadrant where the kink was absent gave no defect indications in the ultrasonic scan.

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*The source strength was about 32 curies, and a focal distance of 20 inches was used. Exposure time was 7 1/2 minutes, with kodak type AA film and lead screens, front and back.

**The test instrument was a Sperry Model UM-721 Reflectoscope, using a 2.25 mHz, 45° transducer. The unit was calibrated on the through crack.

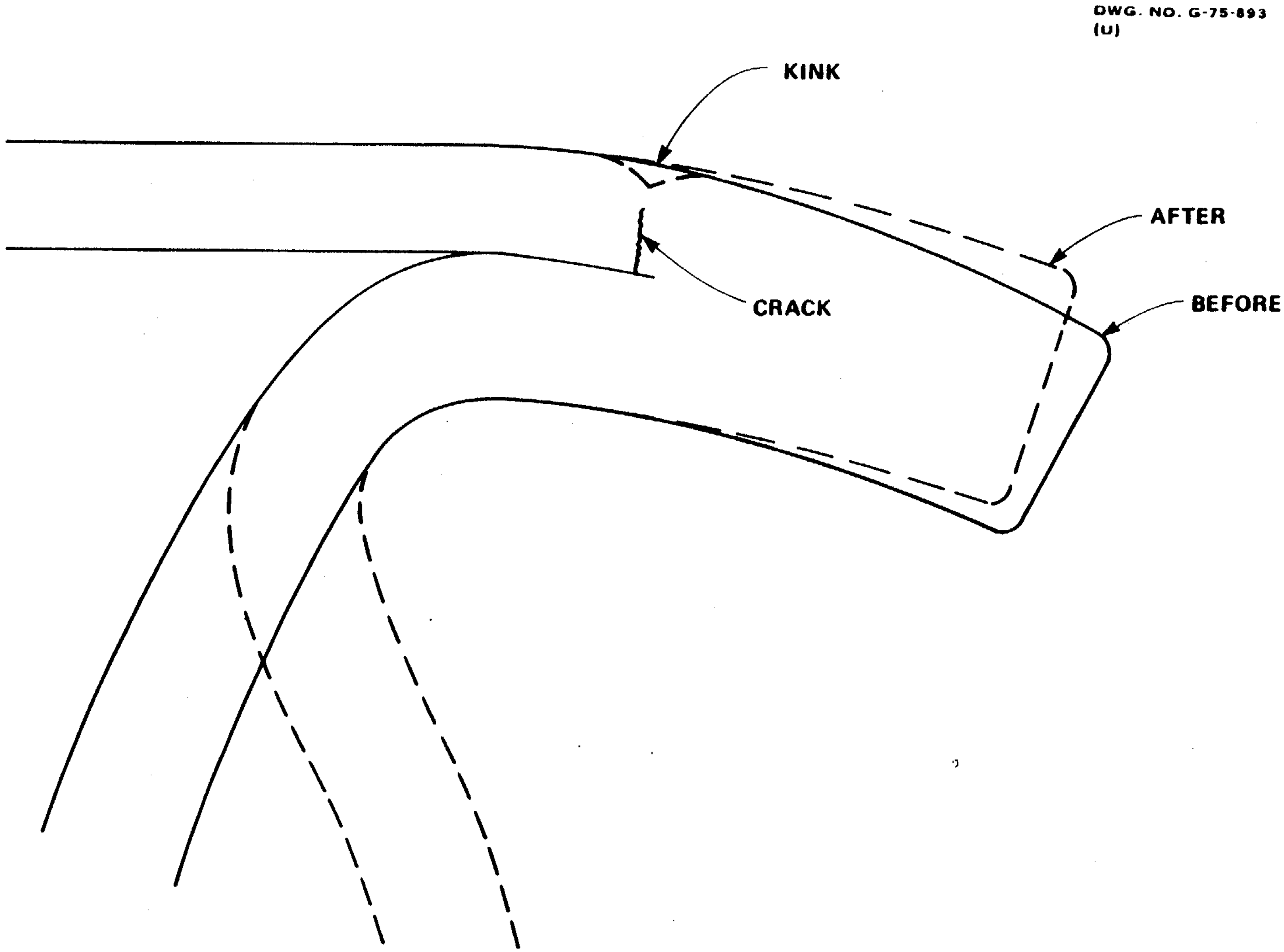

Figure 1

DEFORMATION AND CRACK LOCATION AT PLUG END OF CYLINDER

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(U)

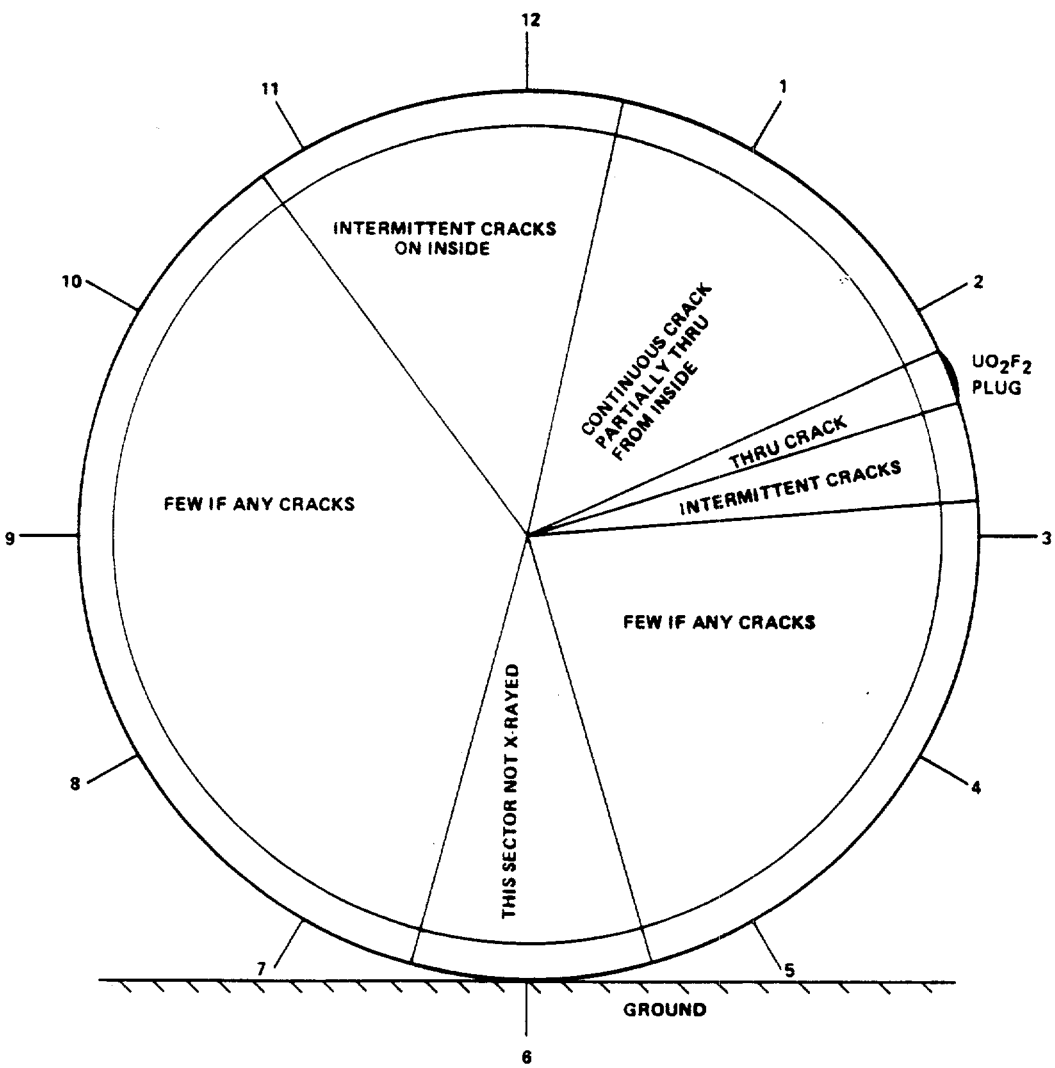

Figure 2

CRACK LOCATIONS AT PLUG END OF CYLINDER AS DETERMINED BY RADIOGRAPHIC EXAMINATION

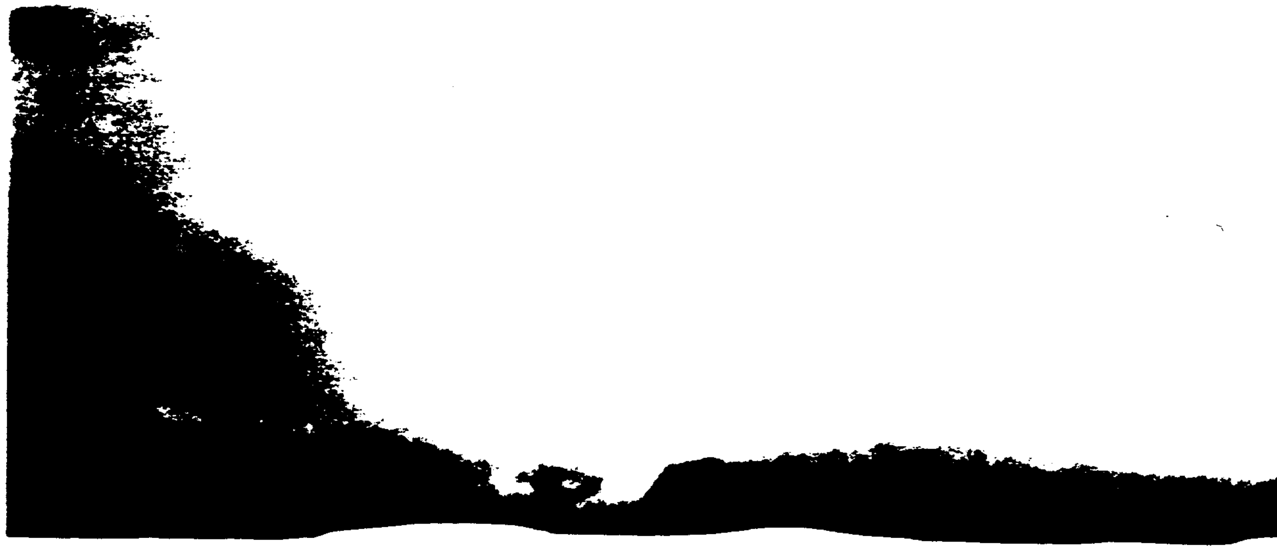

FIGURE 3: Contact Print of Radiograph in Leak Area

The crack is continuous over the left half of the radiograph. The view is radially inward through the flange, with the cylinder end being at the top of the print.

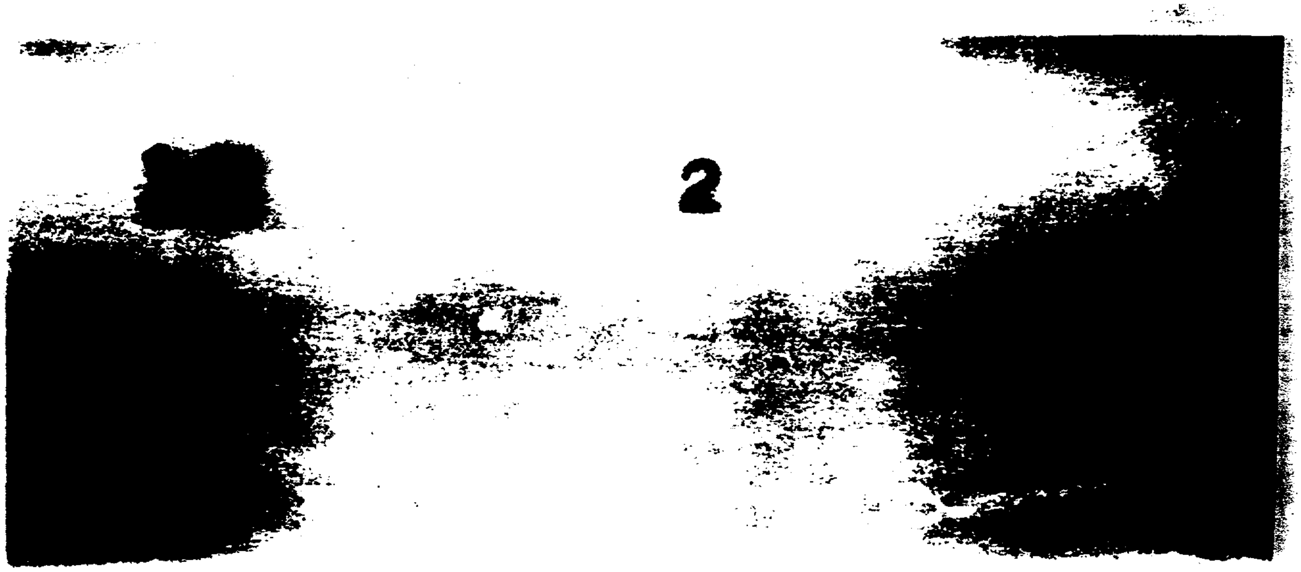

FIGURE 4: Crack in Flange at Top of Cylinder

FIGURE 5: Flange Crack in Area Opposite Leak

FIGURE 6: Flange View at Valve End of Cylinder

No indications of cracking were seen at this end.

Dimensional Inspection Data

Dimensional measurements of length and diameter were taken on Nukem Cylinder No. 8 following the incident. Since there were no prior - incident dimensional data on this cylinder, a twin vessel, Nukem No. 1, was measured to provide baseline dimensional data. The dimensional results from these two cylinders are shown in Figures 7 and 8. Diameter measurements were made with a "pi" tape at the same locations on each cylinder and the overall lengths were measured with a graduated tape. The overall length of Nukem No. 8 was measured to the center of the bulged head which protruded 1 5/8 inches beyond the edge of chine on the plug end of the cylinder. The bulge on the valve end was nearly flush with the chine. The bulging of the two heads was nearly equal since the chine on the valve end was approximately 1 1/8 inches longer than on the plug end. This observation was confirmed on Nukem No. 1, on another twin Nukem cylinder, and on six other random cylinders nearby. The dimensional data indicate that cylinder Nukem No. 1 was near the drawing nominal dimensions except for the difference in chine length. Using the Nukem No. 1 as a base, the average diametrical increase of Nukem No. 8 was 0.66 inches. The length increase due to axial stretching of the cylinder wall was not directly measurable however it was estimated to be no more than 1/2 inch, assuming that the heads bulged symmetrically.

Calculated Volume Change for Nukem No. 8

The increase in internal volume of Nukem Cylinder No. 8 was calculated from post incident dimensional data on the incident cylinder using the dimensions of Nukem Cylinder No. 1 as a base for the estimate. For this purpose, the cylinder geometry was idealized as shown in Figure 9 and geometrical details near the joints and on the bulged heads were ignored. The cylinder interior was assumed to be bounded by a right circular cylinder with concave spherical heads before the incident and with convex spherical heads afterwards. The calculation of the initial volume is as follows:

$$\text{Volume of Spherical Head} = \frac{\pi}{3}(3.749)^2\,(3 \times 30 - 3.749) = 1269.5 \text{ in}^3$$

$$\text{Volume of Cylindrical Part} = \frac{\pi(29.044)}{4} \times 71.686 = 47{,}493.8 \text{ in}^3$$

$$\text{Internal Volume} = V_o = 47{,}493.8 - 2(1269.5) = 44{,}954.8 \text{ in}^3$$

$$\text{or } V_o = 26.02 \text{ ft}^3$$

Similarly, the final volume is estimated as follows:

$$\text{Volume of Spherical Head} = \frac{\pi}{3}(4.970)^2 \times (3 \times 24.679 - 4.970) = 1{,}786.5 \text{ in}^3$$

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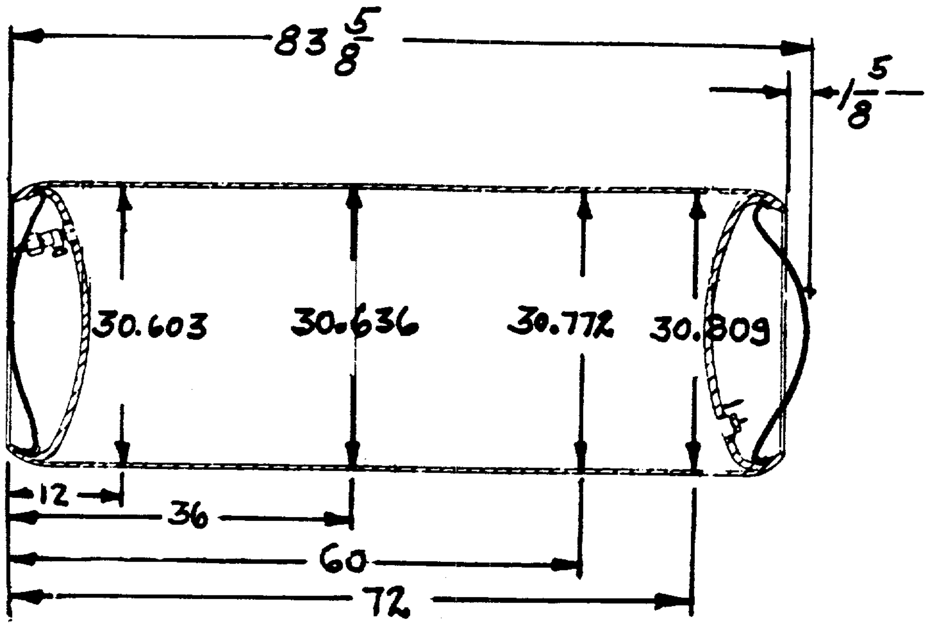

Figure 7
DIMENSIONAL DATA NUKEM NO. 8

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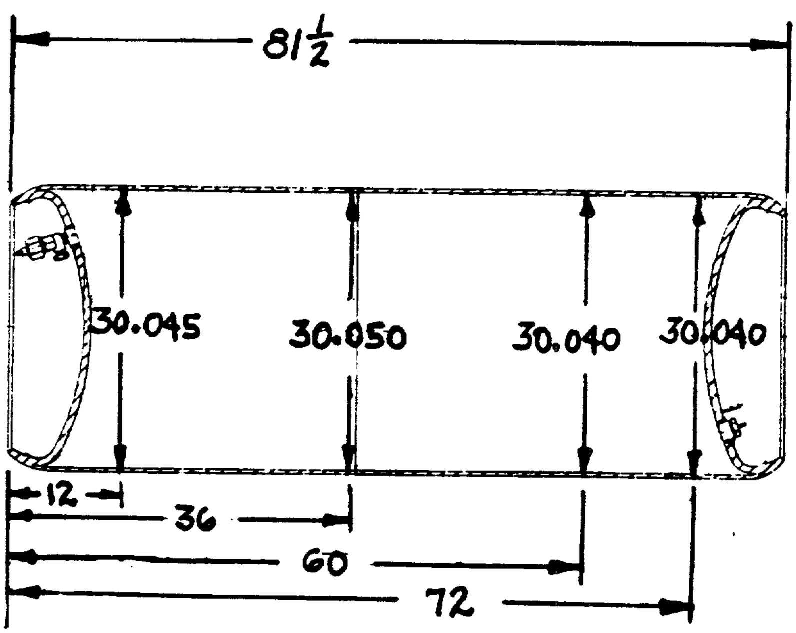

Figure 8
DIMENSIONAL DATA NUKEM NO. 1

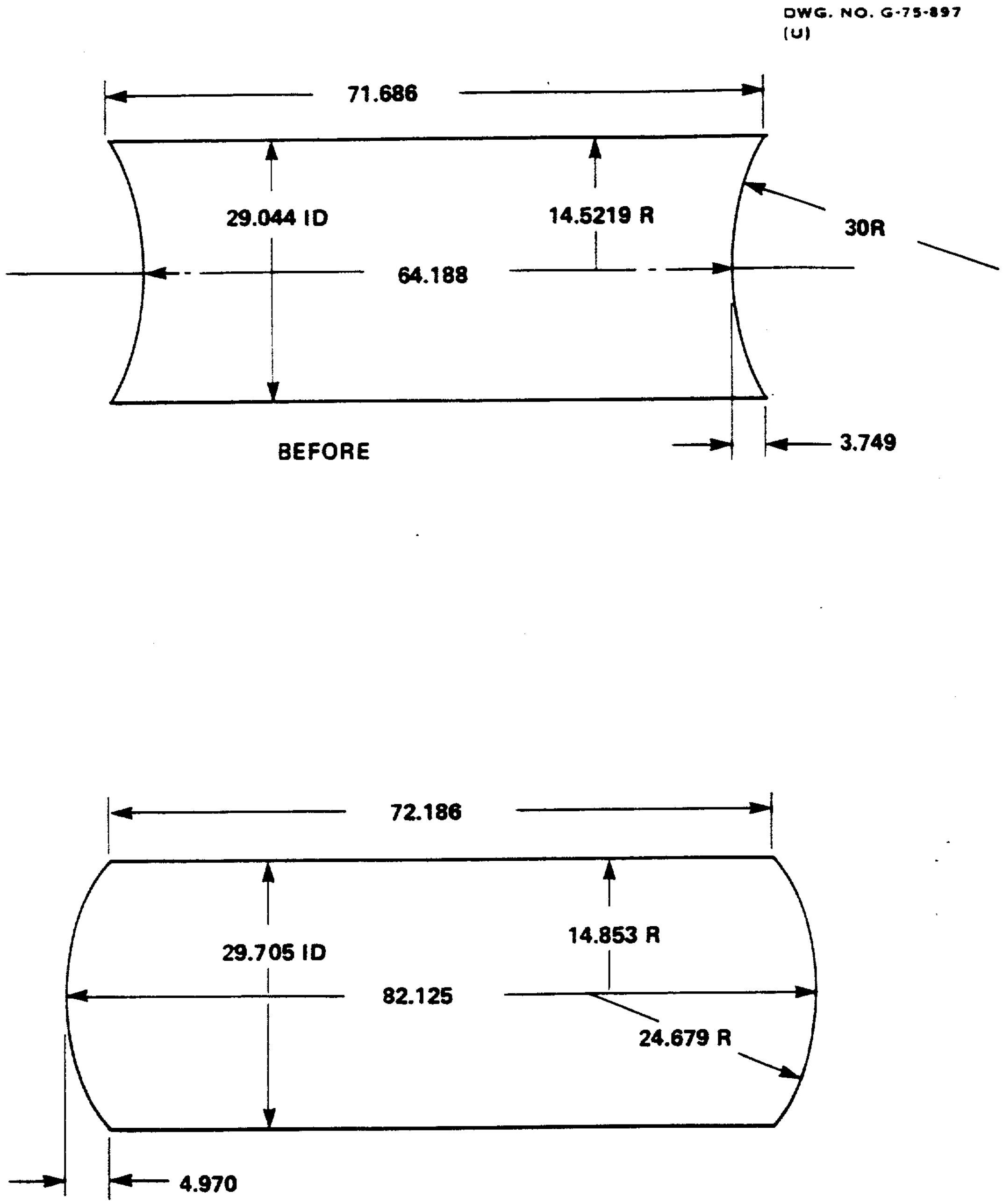

Figure 9
IDEALIZED CYLINDER GEOMETRIES

Volume of Cylindrical Part = $\frac{\pi(20.705)}{4}$ x 72.186 = 50,026.7 in^3

Internal Volume = V_f = 50,026.7 + 2(1,786.5) = 53,599.7 in^3

or V_f = 31.02 ft^3

The calculated volume change is thus:

$V_f - V_o$ = 8,644.9 in^3 = 5.00 ft^3

The percentage increase in volume is calculated as:

$$\Delta V(\%) = \left(\frac{V_f - V_o}{V_o}\right) \times 100 = 19.2\%$$

Of this increase, the head reversal accounts for about 3.5 ft^3 of the total increase.

Estimate of Energy Expended (Volume Change Method)

An estimate of the energy imparted to the cylinder was obtained by calculating the work of expansion of the internal pressure on the volume change of the cylinder. To simplify the calculation, it was assumed that a constant maximum pressure works through the entire volume change. This calculation ignores the elastic expansion of the cylinder as well as the details of the pressure versus volume relationship. It is assumed here that all volume change is a result of plastic deformation in the cylinder. If one assumes that a maximum pressure of 1000 psi acts on a volume change of 8,645 in^3*, the energy is

$$E = P\Delta V = (1000)(8{,}645) = 8.645 \times 10^6 \text{ in-lbf}$$

Or E = 720,417 ft-lbf

Or E = 976,754 joules

Or E = 233.5 kilocalorie (thermochemical)

Damage to Concrete Pad

The sudden change in diameter of the cylinder (particularly due to rapid head reversal) resulted in rapid depression of the surface of the concrete pad directly beneath each end of the cylinder. The damage is shown in Figures 10 through 13. In addition to the two depressions, a crack was formed which ran from the west wall of K-1423 through the centers of both depressions and beyond to an expansion joint in the pad. Measurements were taken of the location and depth profile of the depressions as shown in Figure 14. The distance between the points of maximum depth of penetration (75 3/8 in.) is equal to the distance between the head-to-cylinder knuckles

*See "calculated Volume Change for Nukem No. 8."

PHOTO NO. PH-75-2858

FIGURE 10: Damage to Concrete Pad Caused by Cylinder Reaction

The explosive reaction crushed areas under each end of the cylinder and cracked the full width of the reinforced concrete pad.

PHOTO NO. PH-75-2860

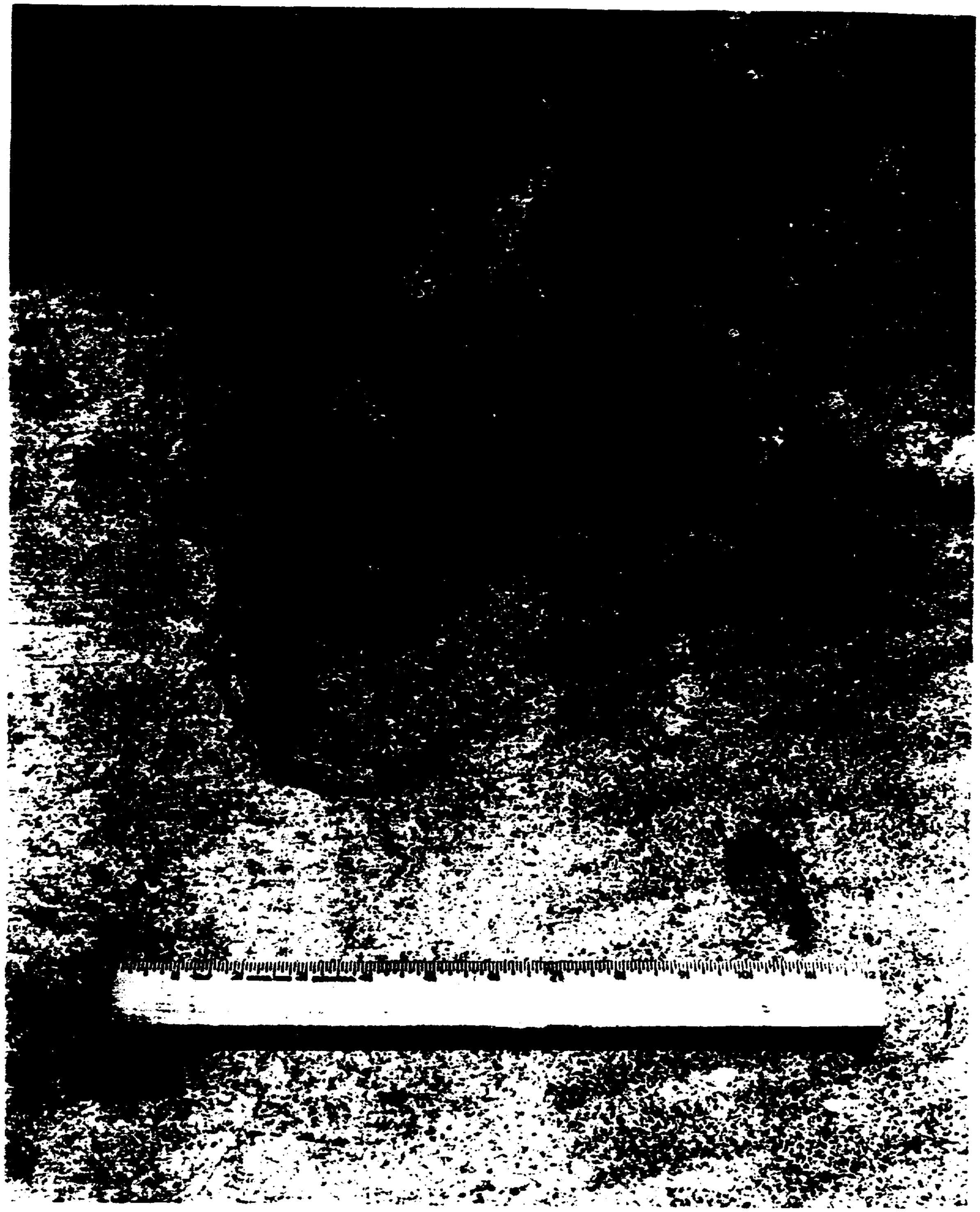

FIGURE 11: Valve-End Depression in Concrete Pad

PHOTO NO. PH-75-2859

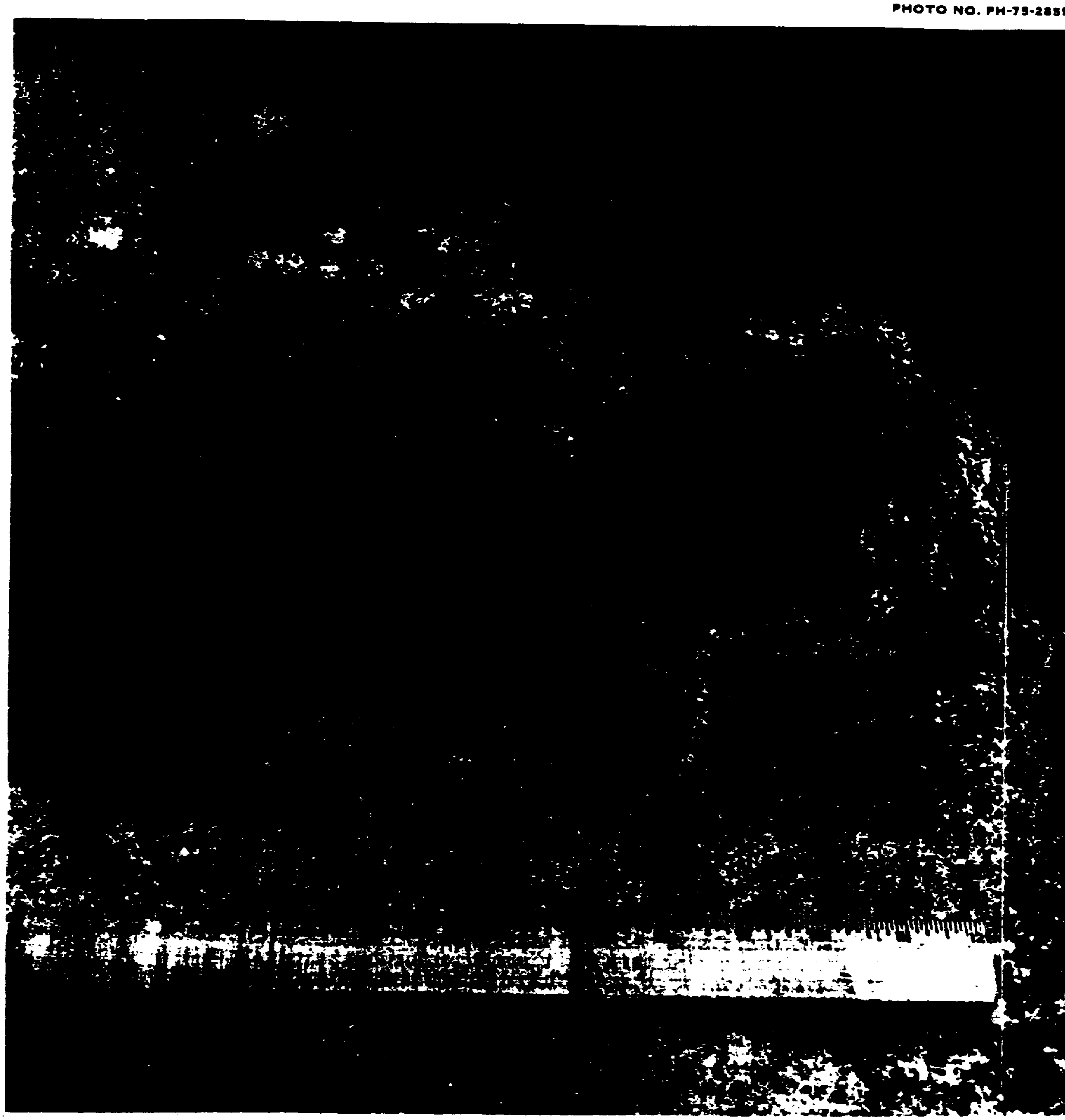

FIGURE 12: Plug-End Depression in Concrete Pad

The deepest part of the damaged area measured 0.3 inch with an average depth of the 9 x 13 inch damaged area being about one tenth of an inch.

PHOTO NO. PH-75-2861

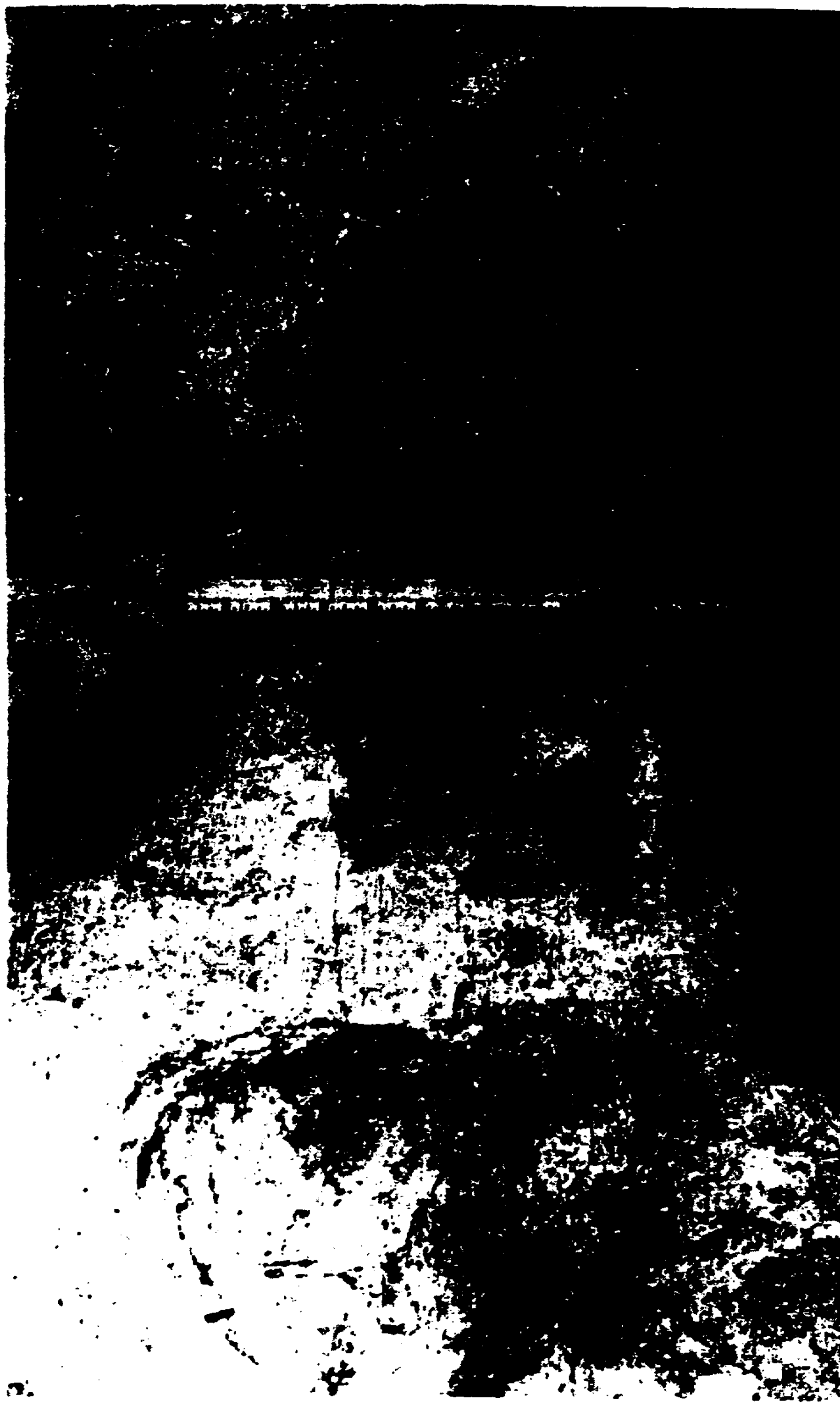

FIGURE 13: Crack in Concrete Pad

The crack extended through the full width of the reinforced concrete pad. Shown here is the 4-foot segment from the building wall through the near (valve end) depression.

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(U)

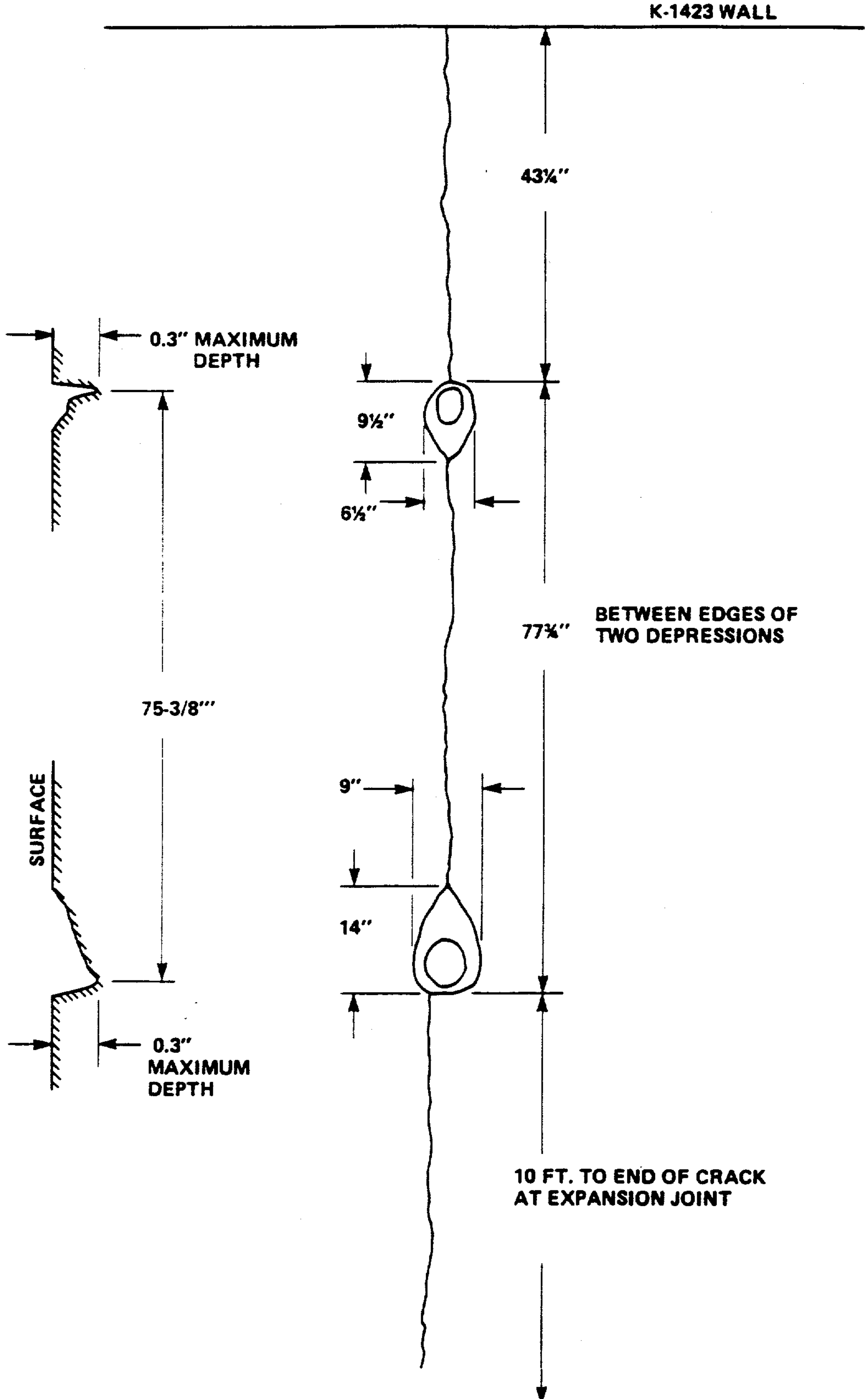

Figure 14

SCHEMATIC ILLUSTRATION OF DAMAGE TO CONCRETE PAD on the 30A type cylinder. This observation suggests that the concrete damage resulted from a rapid increase in diameter (local bulge) at the joints between the heads and cylinder which occurred when the heads reversed. The greater extent of the concrete damage on the plug end of the cylinder suggests a greater increase in diameter on that end and this was confirmed by the post-incident diameter measurements. This collective evidence indicates a greater internal explosive pressure at the plug end which explains why the cylinder wall cracked only on that end.

Energy Exerted to Concrete Pad

An estimate of the energy imparted to the concrete pad was made by calculating the work of compression required to produce the observed damage. For this calculation, a constant stress equal to the compressive strength of the concrete was assumed to act through the volume produced in the two depressions. The dimensional depth profiles were used to estimate the volume of each depression. The work was calculated for an assumed compressive strength of 3,500 psi as follows:

$$W = 3{,}500 \text{ psi} \times 17 \text{ in}^3 = 59{,}500 \text{ in-lb} = 4{,}958 \text{ ft-lbs}$$

This energy is small in relation to the energy expended in deforming the cylinder.

Estimate of Energy Expended (Plastic Work Method)

The energy imparted to the cylinder was also estimated by calculating the plastic work required to permanently deform the cylinder to its final configuration. To simplify the calculation, the material of the cylinder was assumed to behave as a perfectly plastic solid thus ignoring elastic strains and work hardening effects. The value of the yield strength of this idealized material was set equal to the typical yield strength for ASTM-A285-Grade A steel (30,000 psi). For the sake of the analysis, the cylinder was assumed to be fully yielded, with the stresses everywhere being at the yield point. The plastic strains were computed from the dimensional changes as determined from dimensional data* from Nukem Cylinders Nos. 1 and 8. The plastic work was calculated using the equation

$$W_p = (\sigma_y)(\varepsilon_p)(V)$$

where σ_p = yield stress = 30,000 psi

ε_p = plastic strain

V = volume of material

*See "Dimensional Inspection Data"

The plastic work was calculated separtely for each section of the cylinder shell, for each strain direction (i.e., hoop or axial) and for each mode of loading (i.e., tension or bending). The strain components calculated and the equations used are summarized in Table 1 along with the calculated values.

TABLE 1

Strain Calculations

Component	Equation
Hoop Strain in Cylinder	$\varepsilon H = \dfrac{D_f - D_o}{D_o} = 0.22373^{**}$
Axial Strain in Cylinder	$\varepsilon_A \quad \dfrac{L_f - L_o}{} = 0.006975$
Bending Strain in Head	$\varepsilon_B = \frac{t}{2}\left(\frac{1}{R_o} + \frac{1}{R_f}\right) = 0.027626$
Stretching Strain in Head	$\varepsilon_S = \dfrac{C_f - C_o}{C_o} = 0.047554$

Where D_f = final diameter of cylinder = 30.205 in. *

D_o = initial diameter of cylinder = 29.544 in.

L_f = final length of cylinder = 72.1855 in.

L_o = initial length of cylinder = 71.6855 in.

t = thickness of head = 0.75 in.

R_f = final radius of head = 25.0536 in.

R_o = initial radius of head = 29.625 in.

C_f = final arc length of head = 31.7986 in.

C_o = initial arc length of head = 30.3551

*The number of significant digits shown in all calculated results do not reflect the accuracy of that particular result.

The plastic work for each of the strain components listed in Table 1 are calculated as follows:

Hoop Plastic Work on Cylinder = 30,000 x 0.022373 x 3,326.7525
= 2.2329×10^6 in-lb = 186,073.6 ft-lb

Axial Plastic Work on Cylinder = 30,000 x 0.006975 x 3,326.7525
= 6.9612×10^5 in-lb = 58,010.2 ft-lb

Bending Plastic Work on Head = 4 x 30,000 x 0.027626 x 542.7692
= 1.7993×10^6 in-lb = 149,945.4 ft-lb

Stretching Plastic Work on Head = 4 x 30,000 x 0.047554 x 542.7692
= 3.0973×10^6 in-lb = 258,108.5 ft-lb

The total plastic work is the sum of the separate parts as follows:

Hoop, Cylinder	= 186,073.6 ft-lb
Axial, Cylinder	= 58,010.2 ft-lb
Bend, Head	= 149,945.4 ft-lb
Stretch, Head	= 258,108.5 ft-lb
Total Plastic Work	= 652,137.7 ft-lb

To this total one must add the work done on the concrete pad* which is estimated at 4,958.3 ft-lb. The total work due to permanent deformation of the cylinder and the concrete is thus:

W = 657,096 ft-lb

This total permanent work estimate is fairly close to the energy estimated by the volume expansion method. Ideally, the work done by the expanding gases in the cylinder would be equal to the sum of the energies expended through all dissipative mechanisms such as plastic work in the cylinder, elastic deformation converted to heat, acoustic energy, kinetic energy, and work expended in the concrete. Thus, it is not surprising that the energy as estimated by volume change is larger than the estimated work of permanent deformation in the cylinder and concrete pad.

Collapse Pressure of Heads of 30A Cylinder

Two hydrostatic tests of two 30A type cylinders[1] indicated cylinder rupture at 1050 to 1250 psi pressure. A earlier report[2] indicated the start of cylinder head reversal at as low as 600 psi pressure. An analysis of the head reversal process was made to determine the most

*See "Energy Exerted to Concrete".

likely mode of collapse of the heads and to determine the range of pressure within which it would occur. The analysis consisted of two parts, namely an elastic buckling analysis and a plastic limit analysis. The elastic buckling check revealed the design to be stable and thus the heads could not reverse suddenly in a elastic fashion (snap-through type buckling). The plastic limit analysis determines the pressure range over which plastic buckling could occur. For the analysis, the cylinder head was treated as a spherical cap with clamped edges. A solution to this problem was found in Hodge's book[3]. For the dimensions of the 30A head and an assumed yield strength of 24,000 psi (ASTM-A-285 Grade A), the solution predicted lower and upper bounds on the collapse pressure P as follows:

$$1215 \text{ psi} < P < 1330 \text{ psi}$$

Complete collapse of the head would be expected somewhere between these bounds. The start of head reversal would occur when yielding first occurs at the knuckle transition between the head and cylinder[2]. It is likely that this "first" yielding could occur below 900 psi and well below the collapse pressure. Also the plastic limit analysis did not account for the rotation of the transition joint between the cylinder head and wall. It is likely that the collapse pressure would be strongly influenced by the degree of fixity of this joint.

References

1 Ziehlke, K. T., "UF_6 Shipping Cylinders: Hydraulic Tests:, Letter to A. J. Mallett, February 22, 1965. (K-L-1960).

2 Kirstowsky, E. C., and Hain, E. W. R., "Strength Analysis of Type D Chlorine Head, Serial No. D-36337; C&CC No. 203258", Letter to J. A. Marshall, September 13, 1951.

3 Plastic Analysis of Structures, P. G. Hodge, Jr., McGraw Hill, 1959.

APPENDIX F

ANALYTICAL RESULTS

APPENDIX F
ANALYTICAL RESULTS
E. J. Barber

Examination of Samples from Parent Cylinder

Analysis of the original liquid sample from the parent cylinder shows that the material met in every respect the specifications which have been recommended for feed to the gaseous diffusion plant (see Table F-1). Gas samples taken from the cooled parent cylinder after removal of 3/5 of its contents show the same impurities in approximately the same concentrations as the gas over a solidified sample drawn as liquid from the full cylinder. As a result of these studies, it is concluded that there is nothing in the analyses of the material in the parent cylinder either before or after the incident which indicates anything other than that the parent cylinder contained a quality product and that there were no impurities in the parent cylinder in sufficient quantity to have caused the incident with Nukem-8.

Examination of Material in the K-1423 Cold Trap and Alumina Traps

Since the Nukem-8 cylinder was bled once to the cold trap in K-1423 during filling, there was a limited possibility of detecting a gaseous reaction product in the gas over the solid UF_6 in the cold trap and somewhat lesser probability of its retention by physical adsorption on the alumina traps. No gas not normally encountered over the condensed UF_6 in the cold traps was detected. Since the probability of detecting any of the reaction product gases on the alumina bed was even lower, these were not analyzed except for accountability purposes.

Examination of Frozen Cake Containing Hydrolyzed UF_6

Since this material was hydrolyzed as it was leaking from the cylinder valve, it was thought that it might contain some of the reaction products in addition to hydrated UO_2F_2; if a nuclear excursion had been involved, fission products would be present. The analysis indicated the absence of fission products expected from a nuclear excursion. The gaseous products trapped in the ice were those which would have been found had a similar operation been performed on an undamaged cylinder and included the following: CO_2, 72.4%; N_2, 14.5%; O_2, 8.7%; acetone, 3.4%; Cl_2, 4.0%. The trace of acetone was probably accumulated during storage in a freezer in a laboratory room in which acetone is employed as a cleaning solvent.

Examination of Gases from Nukem-8 After Containment and Relocation at K-29

The mass spectrometric analysis of the gas removed from the pigtail before evacuation and similar samples taken later following initiation of feed to the cascade show the presence of significant quantities of carbon tetrafluoride, >40%, and lesser amounts of trifluoromethane, difluoromethane, hexafluoroethane, and partially fluorinated ethanes. An infrared scan

TABLE F-1

URANIUM HEXAFLUORIDE ANALYSES

Property	Specification Limits, Minimum	Units	Basis	Sample Numbers: 964955 Cylinder Numbers: 9634
Uranium Hexafluoride	99.5	Percent	Weight	99.97
Uranium		Percent	Weight	67.60
Cylinder vapor pressure at 200°F	Maximum 75	psia	Measured on: Shipping Cylinder	< 75
Hydrocarbons, chlorocarbons, partially substituted halohydro-carbons	0.01	Percent	mole	< 0.01
Antimony	1	ppm	U	< 1
Bromine	5	ppm	U	< 1
Chlorine	100	ppm	U	54
Niobium	1	ppm	U	< 0.2
Phosphorus	50	ppm	U	< 20
Ruthenium	1	ppm	U	< 1
Silicon	100	ppm	U	7
Tantalum	1	ppm	U	< 0.5
Titanium	1	ppm	U	< 0.3
Elements forming non-volatile fluorides	300	ppm	U	15
Chromium	1500	ppm	^{235}U	< 152
Molybdenum	200	ppm	^{235}U	< 6
Tungsten	200	ppm	^{235}U	< 15
Vanadium	200	ppm	^{235}U	< 9
Uranium-233	500	ppm	^{235}U	< 100
Uranium-232	0.110	ppm	^{235}U	< 0.02
Boron equivalent cross section	8	ppm	U	< 1.8
Fission product gamma	20	Percent of Aged Natural U		< 5
Fission product beta	10	Percent of Aged Natural U		< 2
Transuranic alpha	1500	d/m/g	U	< 25

of the same gas samples indicated the presence of major amounts of carbon tetrafluoride with lesser amounts of other fluorohydrocarbons. The infrared scan also indicated the presence of small amounts of carbonyl fluoride (COF_2). These fluorocarbon and fluorohydrocarbon materials are normal products of the rapid fluorination of hydrocarbons or other high molecular weight organic materials containing principally methylene ($-CH_2-$) groups.

The gas examined is believed to be typical of that which bled from Nukem-8 cylinder when the original cracked valve was replaced. A significant amount of the carbon tetrafluoride also escaped with the UF_6 and HF at the time of the incident.

Inspection and Sampling of Nukem-8 After Feeding

After the UF_6 had been fed to the cascade and the cylinder and containment vessel weighed at K-1423, the unit was taken to K-1420 for inspection, sampling and final decontamination for metallurgical studies. The cylinder was vented to atmospheric pressure from an initial pressure of about 1 psig without seeing any evidence of HF or UF_6. This is in agreement with the gas analysis of the cylinder which showed 5 ppm U and 515 ppm F. The secondary containment vessel which contained 1 ppm U was then bled down to atmospheric pressure, the secondary containment vessel head removed, the valve to the cylinder closed, and the pigtail removed all without visual evidence of HF or UF_6. The cylinder valve was then opened and removed. A sample of orangish yellow material was removed from the valve port. This proved to be partially hydrolyzed UF_6, a material similar to but not identical with $U_3O_5F_8$. The interior of the cylinder was then viewed using a penlight and the normal inspection probe. The atmosphere in the cylinder was clear with no fog. A powder bed was visible at the bottom of the cylinder along the entire visible length. This powder bed appeared deeper at the cracked end of the cylinder. The surface material on the bed surface nearest the cracked end reflected light better (appeared lighter) than the general powder bed. There was more light absorptive (darker) material on the bed surface out a little way from the cracked end. Lumps of material of a composition different from the general powder bed were noted on the surface of the powder bed. Some solid materials stuck to the walls and ends of the cylinder. These were similar to the lumps in the powder bed. A large hard mass of orange colored material was situated at the position where the cylinder had been breached. This material, similar in appearance to the material removed from the valve, proved also to be partially hydrolyzed UF_6 slightly richer in fluorine content than UO_2F_2.

Upon examination, some of the lumps were pliable and generally black in appearance and others were very hard and olive-green in color at the surface but were black when broken open. A few very hard lumps were black on the surface and all the way through. Samples of the powder were a light olive-green in color and well crystallized. The color fitted the description of one preparation of β-UF_5 given in Katz and Rabinowitch, "The Chemistry of Uranium." Examination by X-ray diffraction indicated the powder to be β-UF_5 with no identifiable second phases; however, later sieving of a large amount of the powder showed about 4% of the powder was a dark brown crystalline material identifiable as U_2F_9. SEM analysis showed technetium to be the only significant impurity in this powder. Radioanalysis of this powder indicated that technetium was present in the parent cylinder at 0.07 ppm and at 1.03 ppm in this residue representing about a 15-fold concentration and indicating, not unexpectedly, that essentially all the technetium remained in the residue since TcF_6 is more reactive than UF_6. Carbon was below the detectable limit in the powder. This result is not too surprising in view of the relatively small amount of carbon reacted with respect to the uranium hexafluoride reduced and the fact that carbon tetrafluoride was the major carbon product. The bulk density of the powder was about 2.2 g/ml and the X-ray crystallographic density is 6.5 g/ml.

The lumpy materials provided more information about the reaction. The pliable material proved to be a fluorinated carbonaceous material with a detectable β-UF_5 content. The hard lumps which were green on the surface but black inside contained carbon at the detectable limit along with UF_4, the primary material in lumps fitting this description. Other hard lumps contained mixtures of U_2F_9, UF_4, and β-UF_5. The existence of these hard lumps containing carbon residues indicate the existence of a localized (limited) high temperature zone in the Nukem-8 cylinder. The large rubbery black lumps indicate the presence of a high molecular weight hydrocarbon, most likely oil saturated with UF_6, as a separate phase at the time of the explosive reaction.

The Chemistry of the Incident

Two laboratory studies of the reaction of vacuum pump oil from the valve shop with gaseous UF_6 are also pertinent to understanding what happened. In the first of these, gaseous UF_6 was admitted to a reactor containing a 2-inch diameter aluminum dish with a thin layer of oil at room temperature and heated over a 70-minute period to 200°F. Only a slight blackening of the oil at the edge of the dish and an inconspicuous coating on the oil surface were noted. The study was repeated using UF_6 gas at 500 torr and holding it in contact with pump oil at 200°F for two hours with similar results. The logical conclusion is that the reaction between gaseous UF_6 and vacuum pump oil at 200°F, about 20° higher than the temperature of a normally filled 2.5-ton cylinder, is quite slow.

The laboratory studies indicate that in the slow reaction favored by gas-liquid contact that fluorination of the oil with formation of UF_5 is favored. The gases from the damaged cylinder, Nukem-8, indicate that the explosive reaction may favor formation of gaseous fluorocarbons or fluorohydrocarbons. In either case, slow or fast reaction, the principal reduction product is uranium pentafluoride; while UF_4 and U_2F_9 have been identified in the lumpy products, these occurred as minor fractions of a percent of the residual material. That more energy is released by forming carbon tetrafluoride is evident from the comparison of the standard heats of reaction; i.e., for the reaction

$$\{CH_2\}_{(\ell)} + 2UF_{6(\ell)} \rightarrow 2HF_{(g)} + \{C\}_{(s)} + 2UF_{5(\ell)} \qquad (1)$$

$$\Delta H^\circ_{298} = -44 \text{ kcal}/\{CH_2\} \text{ or } 3.14 \text{ kcal/g oil}$$

for the reaction

$$\{CH_2\}_{(\ell)} + 4UF_{6(\ell)} \quad 2HF_{(g)} + 4UF_{5(s)} + \{CF_2\}_{(\ell)} \qquad (2)$$

$$\Delta H^\circ_{298} = -56.7 \text{ kcal}/\{CH_2\} \text{ or } 4.05 \text{ kcal/g oil}$$

and for the reaction

$$\{CH_2\}_{(\ell)} + 6UF_{6(\ell)} \rightarrow 2HF_{(g)} + 6UF_{5(s)} + CF_{4(g)} \qquad (3)$$

$$\Delta H^\circ_{298} = -118 \text{ kcal}/\{CH_2\} \text{ or } 8.41 \text{ kcal/g oil}$$

or 15,100 BTU/lb oil reacted.

Assuming reaction (3) is followed and the total energy involved is as calculated by Blake and Ziehlke to be about 230 kcal based on the cylinder volume expansion, a total of 230 kcal/8.41 kcal/g oil $\cong$ 27 g oil reacted (about 30 cc) would be required to produce the energy needed, provided all the energy released were employed in the explosion process. Because such efficiency of energy utilization is, in a practical case, not realizable, this is the minimum requirement. It is known that much more oil was involved since about 300 lb of UF_5 were recovered in the reaction residue. Since some carbonaceous residue remained, reaction (3) provides a lower bound as to the amount of hydrocarbon required which is 970 g (2.3 lbs) or a little more than a liter. Equation (2) probably provides an upper bound since conversion of $\{CF_2\}$ groups to CF_4 spontaneously results in liberating half the carbon as elemental carbon. This quantity is 1450 g (3.2 lbs) of oil or about 1.6 liters. Based on pump failure simulation studies reported elsewhere, these quantities are reasonable.

Returning to Equation (3) and assuming an oil containing 40 carbons/molecule, the mole fraction of UF_6 in the exploding mixture would have to be greater than 0.99 if only the minimum required oil were present. The presence of significant amounts of incompletely fluorinated hydro-fluorocarbon molecules suggests that the reaction occurred in a hydro-carbon rich phase which in turn indicates that it is unlikely UF_6 is soluble to so great an extent in the oil. For these reasons, a significantly greater quantity of oil is likely necessary to contain the dissolved UF_6 in sufficient amount to produce the required energy release as indeed is the case as indicated by the calculations of the previous paragraph. That temperatures high enough to fuse UF_4 particles existed in a limited zone in the cylinder at least momentarily is indicated by the analyses of the lumpy materials.

To more firmly establish the explosion mechanism, the possibility of the reaction raising the temperature sufficiently to hydrostatically rupture the cylinder has been investigated. The heat capacity of the cylinder and contents is about 370 kcal/°C (205 kcal/°F). Assuming the reaction goes by Equation (3), the 304 lbs of UF_5 correspond to the generation of 8200 kcal which will raise the cylinder temperature under adiabatic conditions 22°C or 40°F which will not be sufficient to fill the void space by thermal expansion. Assuming 1250 psi are required to rupture the cylinder and the quantities of HF and CF_4 generated to produce the 304 lbs of UF_5, the expected solubility based on experiment for HF and on Raoult's law solubility for CF_4 are sufficiently high to predict accomodation in the liquid of sufficient quantities of HF and CF_4 to prevent overpressuring the cylinder, unless a reaction of explosive violence occurs.

The Scenario of the Incident

A 30A type cylinder (Nukem-8) containing between 1.0 and 1.6 liter of hydrocarbon oil was connected for filling with liquid UF_6 by transfer from a parent cylinder at about 205°F. As the liquid UF_6 entered the cylinder it first froze until the temperature of the cylinder rose to 64°C after which all the frozen UF_6 melted. The oil being lighter than UF_6 rose to the surface where it floated as a thin layer on the liquid UF_6 which was slowly reacting with the oil, but not as rapidly as the UF_6 was diffusing into the oil phase. The oil phase was concentrated near the rear of the cylinder by the inrushing UF_6. Gradually the reacting quantities increased not so much because of increasing reaction rate but rather because of increasingly greater quantities of UF_6 dissolved in the oil phase. After about an hour the reaction had produced sufficient gas to prevent fully filling the cylinder. The cylinder was given the burping customary when filling difficulties are encountered and the filling resumed at a slower rate. By this time the reacting oil was becoming less resistant to additional attach as is typical of such residues and the rate was picking up; however, the cylinder also reached its fill limit, was disconnected, and weighed. During this period the reaction was increasing in intensity so that, as the cylinder was set down, the critical velocity was reached and the UF_6 saturated oil layer explosively reacted as a result of the culmination of a chain reaction involving a free radical transfer process and increasing sensitive reaction fragments. Two factors appear to be critical to obtaining the reaction. One factor is achieving a high concentration of UF_6 in the oil phase which for practical considerations means the presence of liquid UF_6; the second factor is the buildup of a concentration of reactive organic fragments in the oil which requires time. It is also possible that sloshing the material around during setting down the cylinders is a contributing, but not a necessary, factor. The explosion reversed the heads of the cylinder, cracking one end and damaging the valve by hammering it against the chine on the other.

APPENDIX G

NUCLEAR CRITICALITY SAFETY CONSIDERATIONS

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NUCLEAR CRITICALITY SAFETY CONSIDERATIONS

H. R. Dyer

Moderation control has long been recognized as an acceptable, single parameter control for maintaining nuclear criticality safety in processing large quantities of low enriched uranium hexafluoride. Adequate controls must be in effect to prevent hydrogenous materials from entering the UF_6 containment system, as well as to prevent escaping UF_6 and reaction products from collecting in unsafe volumes, in the event the containment control is lost. There are elements other than hydrogen capable of moderating neutrons, but hydrogen bearing compounds, such as water and oils, are the most likely to be encountered.

Hydrogenous materials mixed with UF_6 presents two types of hazardous reaction potentials, chemical and nuclear. The chemical reactions are not, per se, the principal concern of criticality safety; however, the circumstances which lead to and the results after the reaction can be important. The nuclear reaction, i.e., criticality, is the concern of all personnel. The potential of an accidental criticality is dependent upon many variables, such as, but not limited to, the quantity and concentration of uranium, uranium isotopic assay, quantity of moderating materials present, size of containment system.

The incident at K-1423 involving the 2.5-ton UF_6 cylinder illustrates all of the above concerns, from a criticality safety standpoint. The violent reaction was obviously the result of a reactive hydrogenous contaminant in the cylinder. Since the "moderation control" safeguard had been breached, one could only speculate on the amount of hydrogenous material present. It was therefore not inconceivable that the reaction could have been nuclear as well as chemical. Since the incident occurred outside the building, and there was a light rain, there was also the potential of introducing water into the cylinder. Fortunately, the steel cylinder had only small external cracks, making the entrance of water highly improbable. Of greater concern was the possibility of the cylinder integrity completely failing and releasing large quantities of UF_6. This possibility, combined with the rainy weather and the presence of near-by storm drains and underground cable tunnels presented additional undesirable potentials for a criticality.

The actions taken by the Criticality Safety Group were principally precautionary. Initially, gamma readings were taken to determine if a criticality had occurred in the cylinder, even though the criticality alarm had not sounded. Since these readings of about 0.1 mr/hr at the surface were the same as other 30A cylinders with similar U-235 enrichments, it was concluded that a criticality had not occurred.

To ensure that the circumstances did not propagate into a more serious criticality potential, the following actions were taken:

1. The cylinder was covered to protect it from inclement weather.

2. The near-by storm drain and three other manhole covers were sealed.

3. Nuclear safe vacuum cleaners were made available.

4. Five-gallon buckets and shovels were made available.

5. The borated water pumper was made available.

6. A check was made on the availability of cadmium nitrate solution (maintained in special storage).

These precautions were in effect until the damaged cylinder had been loaded into the secondary containment vessel and transported to the Feed Point.

Due to the many unknown conditions, such as cylinder integrity, quantity, and composition of contents, amount of UF_6 released, cause of the initial reaction, and the possibility that almost anything could still happen, these precautionary measures were taken to reduce the probability of an excursion to as low as possible.

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